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Metrics for quantifying the circularity of bioplastics: the case of bio-based and biodegradable mulch films

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Abstract

The concept of circularity and its quantification through the Material Circularity Indicator (MCI) is well established for traditional plastic products. In this paper a methodological approach for calculating the circularity of bio-based and biodegradable (BB) products is proposed and applied to BB mulch films. BB products are different from traditional products in as much as they are sourced and regenerated (recycled) not through technical cycles but the biological loop. The suggested method is an adaptation of the MCI where two major changes were made: (i) the mass of the bio-based component corresponds to the recycled material in input and (ii) the mass of the bio-based component leaving the system through composting or biodegradation in soil is accounted as recycled. The modified MCI supports the Eco-design of innovative BB products and allows for the comparison of their circularity taking into account the biological source and the expected end of life process such as biodegradation. To demonstrate the adaptation, the method has been applied to BB mulch film. Results showed that the MCI of a biodegradable mulch film, characterized by an average bio-based feedstock content of 30% is 0.37 ± 0.04 in a 0-1 scale. For BB mulch film, the amount of bio-based feedstock is the most sensitive factor and controls linearly the value of the MCI.

Keywords: circularity indicators, circular economy, bioplastics, biodegradable mulch film, bio-based product, biodegradation

Abbreviations

BB	Biodegradable and bio-based
CE	Circular Economy
d.m.	Dry matter
EMF	Ellen MacArthur Foundation
LCA	Life Cycle Assessment
LDPE	Low-Density Poly-Ethylene
MCI	Material Circularity Indicator
NRF	Non-Restorative Flows
PBAT	Polybutylene adipate terephthalate
PE	Poly-Ethylene
PLA	Polylactic acid

1 Introduction

To overcome today's unsustainable model of 'take-make-dispose' and its related risks such as hikes in raw material prices, pressures on the environment, shortage of global resources and waste sinks, a circular approach needs to be applied. It is a new regenerative economic view, based on a balance between economy, environment and society, a total resource efficiency and a Zero Emission Strategy that aims to maximize products value with zero, or minimal, environmental impact (Ghisellini et al., 2016) . Together with structural changes in environmental legislation, new logistics, technologies and sharing schemes, the Circular Economy (CE) approach which is regenerative by design, aims at closing materials loops, *i.e.* at reducing virgin materials input and waste output.

In December 2015, the European Commission developed an Action Plan for Circular Economy (European Commission, 2015), where plastic was considered a priority to be tackled. In January 2018, an *EU Plastic Strategy* (European Commission, 2018) was adopted, in order to react to the increasing environmental problems concerning plastic production, consumption, use and disposal along the same lines of the CE approach. Two fundamental steps to increase the circularity of different plastic products are (i) the abandonment of fossil fuels, *i.e.* currently 90% of the plastic is produced by virgin petroleum-based feedstock (Ellen MacArthur Foundation, 2017), and (ii) the development of easily recyclable products which are recycled. Today, in EU the share of plastics collected for recycling is 30% while the use of recycled plastics is just 6% (European Commission, 2018).

Biodegradable and bio-based (BB) plastics are spreading across markets (Institute for Bioplastics and Biocomposites, 2018) as a valid contribution to meet CE aims and principles. This is true as long as the supply of renewable raw materials, generally from agriculture, is based on a sustainable approach and the conversion processes along the

supply chain are efficient and highly integrated in a Life Cycle Assessment (LCA) perspective (EPLCA – European Platform on LCA). While traditional plastics can be mechanically recycled or incinerated with energy recovery, BB plastic products offer new recycling routes in waste management, due to their biodegradability. Organic recycling (through composting or anaerobic digestion) or in the case of specific applications such as agricultural mulch films, biodegradation in the environment, offer additional recovery options resulting in less wastes.

Nevertheless, the research and development of innovative products, such as the BB products, implies the development of methodologies and metrics capable of measuring their circularity. Without this it is not possible to achieve measurable results and improving actions, as well as provide unequivocal references for comparisons of products of the same type/category. In 2015 the Material Circularity Indicator (MCI) was developed (Ellen MacArthur Foundation & Granta Design, 2015) which aims to quantify the regeneration of a product's material flow and is considered one of the few, among sixteen CE indexes suiting a micro-scale assessment of circularity at product or company level (Lonca et al., 2018). However, it focuses solely on technical cycles and recycled materials. Furthermore, recovery and recycling through the biological cycle offered by industrial composting, anaerobic digestion or biodegradation in natural environments are not considered as end of life options. In order to apply the MCI system to BB plastic products, the development of an enhanced methodology is necessary.

The approach proposed by the authors allows to quantify the circularity of BB plastic products (*e.g.* starch-based bioplastics) and to make comparisons with equivalent traditional plastic products. To demonstrate the applicability of the proposed method a computational example for mulch film products is provided. In so doing so, the paper aims at contributing to the Eco-design of these innovative products.

1.1 The case study of mulch films

Plastic mulch films represent an important agronomical technique well established for the production of many crops thanks to numerous agronomical advantages such as: increased yield and higher quality of productions (Steinmetz et al., 2016) ; weed control and reduced use of pesticides; early crop production and reduced soil moisture loss (Briassoulis and Giannoulis, 2018). As a consequence, the plastic films consumption has increased year-by-year, reaching a current global market estimated at 1.4 Mt, mainly in Asia (Briassoulis and Giannoulis, 2018; Mormile et al., 2017) , and covering 80,000 km^2 of agricultural surface (0.6% of the global arable land). The mulch film market in Europe is estimated by Agriculture Plastic & Environment and by the European Bioplastic Associations at 76-80 kt. The most used raw material is Poly-Ethylene (PE) in its different forms, due to its processability, chemical resistance, high durability and flexibility (Kasirajan and Ngouajio, 2012).

Despite these benefits, manifold environmental and agronomic problems have been pointed out. After its useful life – which in general does not exceed 1 to 3 months – the mulch film has to be removed and properly disposed of, a time-consuming and costly procedure. The recovered film is usually heavily contaminated with soil and organic residues, making mechanical recycling technically difficult and not a cost-efficient solution (Briassoulis et al., 2018; Figuier, 2016; De Lèpinau, Arbenz, 2016). The most common end of life of collected films in Europe is still landfilling (about 50%), followed by energy recovering and finally mechanical recycling (Le Moine, 2014). Recent Chinese prohibition (January 2018) to import different types of wastes is heavily impacting the European agricultural plastic waste management, highlighting the difficulty in properly recycling this type of plastics (Tamma, 2018). Plastic films may not be properly collected and recycled but disposed of by burning in the field or by uncontrolled landfilling or left directly in the (agricultural) soils, causing serious environmental concerns. An example is the "White pollution" phenomena described in the Xinjiang Autonomous Region (China), in which the residual plastic film can reach 200 kg/ha in the top soil with detrimental effects on soils' quality, health and fertility (Liu, He, & Yan, 2014; Gao *et al.*, 2019; Steinmetz *et al.*, 2016).

As a reaction, there has been significant research into novel materials especially related to biodegradable and bio-based (BB) mulch films, which enable an effective biodegradation in soil and provide comparable agronomical performances (Touchaleaume et al., 2016). The term "bio-mulch film" brings together several types of both bio-based and fossil oil-based biodegradable polymers and blends of them, such as polylactic acid (PLA), polybutylene adipate co-terephthalate (PBAT), starch-based polymer blends or copolymers. They biodegrade when exposed to bioactive environments such as soil and compost (Kasirajan et al., 2012) which means that they can be left *in situ* to be fully biodegraded after being used. However, their biodegradability must be proved by accredited certification bodies and standardized procedures.

The EN 17033:2018 is a new European Norm (standard) concerning "Plastics - Biodegradable mulch films for use in agriculture and horticulture - Requirements and test methods", which sets the necessary tests and limits to define biodegradability, performances and environmental impacts of BB much films. The material is considered completely biodegradable if it achieves a complete biodegradation (absolute or relative to the reference material) in a test period no longer than 24 months (mineralization into CO_2). Additionally, a control of constituents (such as metals) and eco-toxicity testing (acute and chronic toxicity tests on plant growth, earthworm; nitrification inhibition test with soil microorganisms) were required. A certified mulch film guarantees that the product will completely biodegrade in the soil without adversely impacting on the environment.

1.2 Goal of the paper

The goal of the paper is to provide a general and common metric to measure the circularity of a bio-based and biodegradable (BB) product and to apply the methodology at product level to a category of products, namely bio-based and biodegradable mulch films.

2 Materials and Methods

2.1 MCI accounting according to the EMF methodology

The Material Circularity Indicator (MCI), according to the Ellen MacArthur Foundation (EMF) methodology (Ellen MacArthur Foundation & Granta Design, 2015), is a number that can range from 0 (pure linearity) to 1 (pure circularity). A purely linear production provides for the exclusive use of virgin raw materials that turn into waste at the end of the use phase of the product. Vice-versa, pure circularity includes the use of recycled materials and does not produce wastes (regenerative streams). Circularity can be achieved in different ways: as for the purpose of this paper, only recycling will be considered since reuse is not an option for thin biodegradable mulch films. Since the method considers only mass flows, the recycling corresponds to the recovery of materials for the original purpose or for other purposes and excludes energy recovery, considered as a loss of materials equal to landfill disposal. The materials recovered feed back into the process as recycled feedstock.

The MCI methodology differentiates ‘technical cycles’ from ‘biological cycles’, modelling only the former. The first contains products and materials re-entering into the system (market) with the highest possible qualities and for as long as possible (thanks to reuse, repair, refurbishment and recycling) and the latter includes biological materials used in cascade until their restoration into the biosphere and the re-constitution of natural resources.

The material flows associated to the production of a generic technical cycle from non-renewable sources are summarized in Figure 1. The dashed lines indicate that recycled feedstock does not have to be sourced from the same product but can be acquired on the market. With reference to Figure 1, the list of the parameters used in the EMF methodology is reported in Table 1, while the equations relevant for the analysis carried out in this paper are described in the following sections (Table 2, Chapter 2.2).

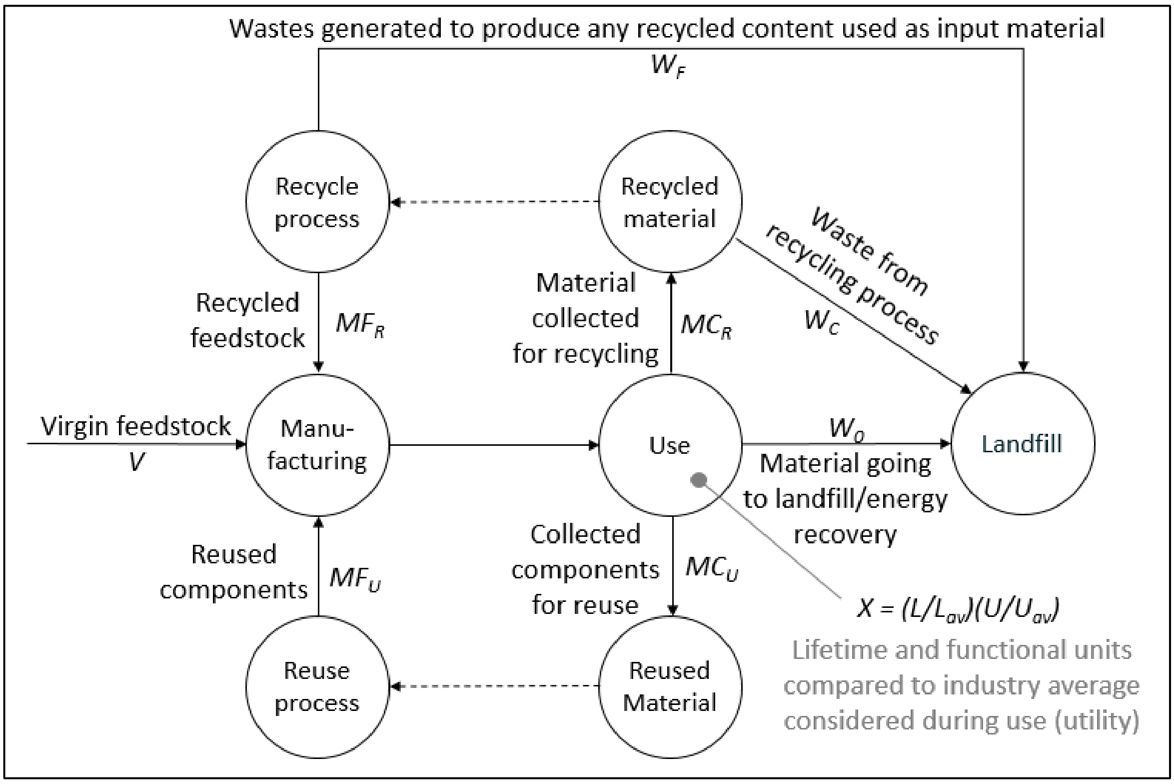

Figure 1: *Diagram of material flows and associated variables of a generic product (modified from Ellen MacArthur Foundation & Granta Design, 2015).*

Table 1: *Parameters and relative definitions used in the EMF methodology.*

Parameter	**Definition**

$\boldsymbol{M}$	Total mass of the product
$\boldsymbol{F_R}$	Fraction of mass of a product's feedstock from recycled sources
$\boldsymbol{F_U}$	Fraction of mass of a product's feedstock from reused sources
$\boldsymbol{V}$	Mass of virgin feedstock used in a product
$\boldsymbol{C_R}$	Fraction of mass of a product being collected to go into a recycling process
$\boldsymbol{C_U}$	Fraction of mass of a product going into component reuse
$\boldsymbol{E_C}$	Efficiency of the recycling process used for the portion collected for recycling
$\boldsymbol{E_F}$	Efficiency of the recycling process used to produce recycled feedstock for a product
$\boldsymbol{W}$	Total mass of unrecoverable waste associated with a product
$\boldsymbol{W_0}$	Mass of unrecoverable waste (landfill, waste to energy and any other type of process where the materials are no longer recoverable)
$\boldsymbol{W_C}$	Mass of unrecoverable waste generated in the process of recycling parts of a product (after use)
$\boldsymbol{W_F}$	Mass of unrecoverable waste generated when producing recycled feedstock for a product
$\boldsymbol{X}$	Utility of a product, calculated as $X = (L/L_{av})(U/U_{av})$

L	Actual average lifetime of a product
L_{av}	Actual average lifetime of an industry-average product of the same type
U	Actual average number of functional units achieved during the use phase of a product
U_{av}	Actual average number of functional units achieved during the use phase of an industry-average product of the same type

The Material Circularity Indicator is determined as follows: , where *LFI* is the Linear Flow Index measuring the flows of virgin materials and unrecoverable wastes associated to the examined product.

A function of the utility, − , is used to correct the *LFI*. The function *F* is chosen in such a way that improvements of the utility of a product (e.g., by using it longer) have the same impact on its MCI as a reuse of components, leading to the same amount of reduction of virgin material use and unrecoverable waste. Setting $a = 0.9$, MCI takes, by convention, the value 0.1 for a fully linear product (*i.e.*, $LFI = 1$) whose utility equals the industry average (*i.e.*, $X = 1$). This leaves some margin to distinguish between processes with a high linearity but different utilities.

2.2 MCI accounting for bio-based and biodegradable (BB) products

To apply the EMF methodology to BB products, formulas and flows (Figure 1 and Figure 2) are adapted as it follows:

1. The fraction of the recycled feedstock, F_R, corresponds to the share of the bio-based feedstock content in the final BB product, $F_{R(i)}$. It is the ratio of the d.m.

amount of bio-based feedstock per d.m. amount of the total mass of BB product (EN 16785-2:2016).

2. The fraction of restorative mass going into a recycling process, C_R, corresponds to the share of bio-based feedstock content in the BB product biologically recovered (*e.g.* through composting) or biodegraded in the natural environment, as it happens for specific applications (e.g. biodegradable mulch film, *etc.*). It is the ratio of the d.m. amount of bio-based feedstock per d.m. amount of the total mass of BB product that is biologically recycled.

The modified scheme is shown in Figure 2. Table 2 lists the formulas as adapted to BB products.

Table 2: *List of formulas as developed by EMF methodology compared to the proposed adaptation to BB products.*

EMF methodology	Adaptation to BB products

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The mass of fossil-based feedstock which may be contained in BB products (*V*) is obtained as a difference of the total mass (*M*) minus the bio-based fraction; in this case the F_R in the EMF methodology corresponds to the sum of the fractions of all the bio-based feedstock/s used in manufacturing the BB product. Therefore, is the total bio-based feedstock mass in the product. In single-use products, such as mulch films, reuse is not considered for BB products, so that $F_U = C_U = 0$.

W_F is the total amount of unrecoverable waste associated to the production of bio-based feedstock used to produce BB products (*i.e.* the amount of uncoverable waste per unit of BB product). Bio-based feedstocks such as starch and PLA generate non-restorative flows which can be quantified. Such unrecoverable waste correspond to $R_{(i)}$, the specific amount of waste generated within cradle-to-gate boundaries per unit of bio-based feedstock going into manufacturing, and it is estimated through LCA studies. Thus all inputs from growth and harvesting phases and the related wastes generated by fertilisers and pesticides are here accounted. $R_{(i)}$ can be easily found in specific literature or life cycle inventories (LCI) present in LCA databases. In the calculation of W_F, also the efficiency of manufacturing process of BB products E_P is considered, as the ratio of the overall bio-based feedstock content in the final BB product to the bio-based feedstock in input to the manufacturing process.

The material flows associated to the production of a generic BB product are summarized in Figure 2.

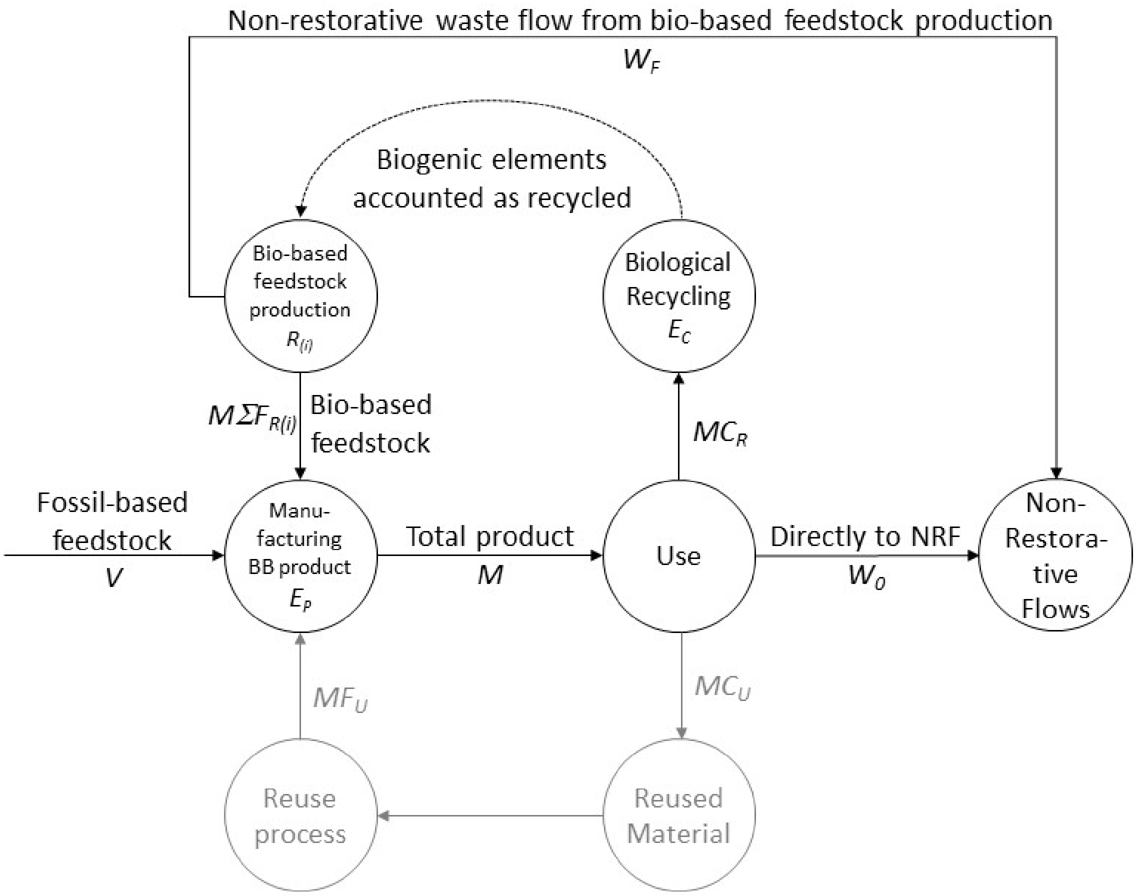

Figure 2: *Description of material flows adaptation to BB products; in this paper, the reuse flow is out of scope ($C_U = F_U = 0$).*

The biodegradation of bio-based feedstock does not imply the generation of waste W_C as it occurs in a standard mechanical recycling process. This implies that C_R and E_C (*i.e.* the efficiency of the biodegradation process) equal to 1. Indeed, a BB raw material, sent to biological treatment (composting) or biodegraded in a natural environment, is fully transformed in its chemical elements (C, H and O mainly) derived from the decomposition of complex molecules (polymers) without the release of waste (Witt et al., 2001; Marten et al., 2003; Eubeler et al., 2010; BASF, 2018; Institute of Bioplastics and Biocomposites, 2018; OWS, 2018; Zumstein et al., 2018). These natural elements return into the environment and are then available in the respective biogeochemical cycles. The (biodegradable) fossil portion behaves as well; consequently, $W_C = 0$.

Nevertheless, the fossil-based feedstock cannot be considered as a regenerative circular feedstock, since it derives from carbon stored for millions of years and extracted by man, not being part of the active and fast biogeochemical carbon cycle. This is accounted in the quantification of W_0, the mass of unrecoverable waste from use (*i.e.* the linear stream going to landfill or incineration, the Non-Restorative Flows, NRF), as , the total amount of fossil-based feedstock.

Since W_F and W_C are associated to complete different processes and W_C is always equal zero, the double counting issue does not occur and the quantification of *W* and *LFI* is modified as reported in Table 2.

2.3 MCI calculation for mulch films: scope, inventory and assumptions

The new formulas reported in Table 2 were applied to a single use product namely a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1.

In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the BB film is assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e. 23% of starch, $F_{(S)}$, and 7% of a bio-based plasticizer, $F_{(BP)}$), while the rest was assumed to consist of fossil feedstock (Figure 3). Since a generalized approach was used and no primary data were implemented, the information were extrapolated from literature; the main characteristics of the two examined products are presented in Table 3.

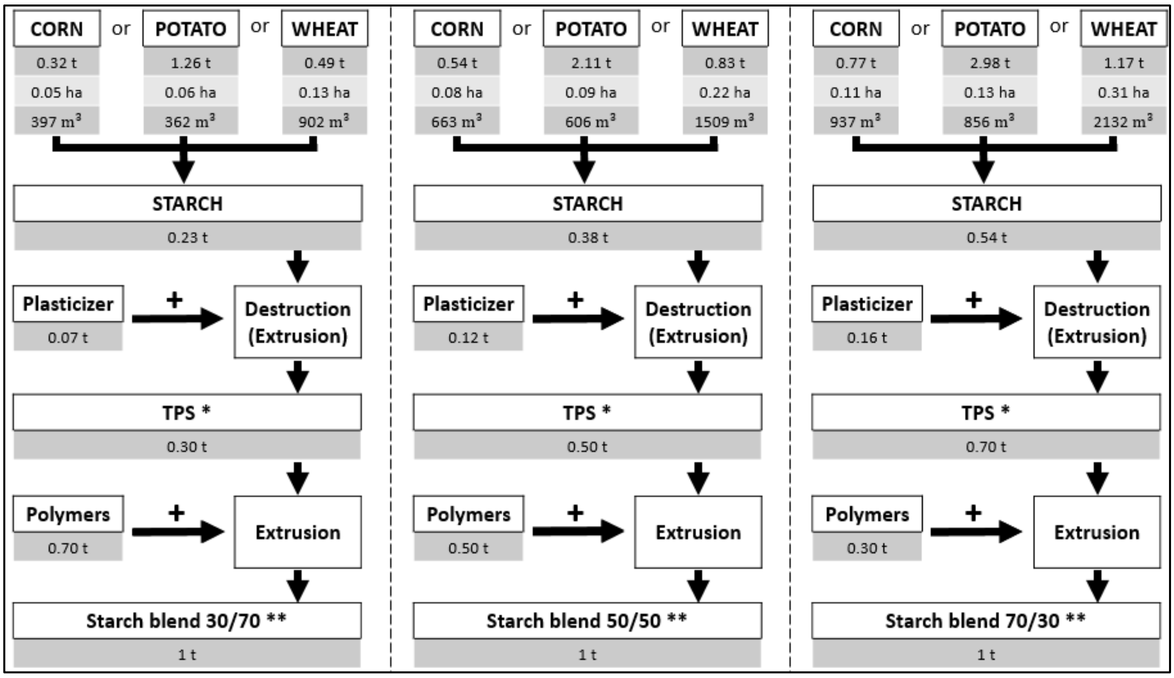

Figure 3: *Examples of starch-based polymers; in this paper, the first option on the left (starch blend 30/70) has been chosen as representative of a BB mulch film. The figure considers a 100%-efficiency in every phase of production, so that the residues are equal to zero; the same assumption is done in this paper. *TPS (Thermoplastic starch), starch content 75%; **Ratio TPS/Polymer; modified from Institute of Bioplastics and Biocomposites, 2018.*

Table 3*: Key features representative of the BB mulch films.*

	BB mulch film
Material	30% bio-based feedstock (23% starch + 7% bio-based plasticizer) + 70% fossil-based feedstock
Thickness (µm)	12
Density (g/cm^3)	1.25
Weight (g/m^2)	15.2
Functional unit (the covering of the agricultural land)	6000 m^2/ha (the actual mulched soil in a hectare is generally equal to the 60% of the total area; Malinconico, 2017)

In the calculation of MCI for the BB mulch film, the adapted formulas were used together with assumptions. As stated before, BB mulch films are blends of bio-based and fossil based feedstocks (in the specific case, 30% and 70% respectively). Unlike the LDPE mulch film that has to be removed and disposed of, the BB mulch film is left in soil where it undergoes an ultimate biodegradation (so that $C_R = 1$) with no waste (so that $E_C = 1$), in respect of the specific standard EN 17033:2018. As a result of polymers' decomposition, the derived (biogenic) C, H and O finally return into biosphere (atmosphere, microorganism biomass, organic material pool), and back into biogeochemical cycles in a relatively short time ("Biogenic elements accounted as recycled" in Figure 2), with the exception of humified compounds. Actually, also C, H and O deriving from fossil-based sources undergo biodegradation but they are not considered as a regenerative flow ("Waste from non-restorative flow" in *Figure 2*) and their "wastes" are indeed calculated in W_0.

Applying a conservative approach, W_F, the waste generated by the production of each bio-based feedstock, is quantified considering a "cradle to gate" LCA study. The estimated solid wastes $R_{(i)}$ for the presented case study are related to the production of starch ($F_{(S)}$), with an amount $R_{(S)}$ of 0.014 kg of waste per kg of renewable feedstock (source: personal communication A. Novelli), and to the production of the bio-based plasticizer ($F_{(BP)}$), with $R_{(BP)}$ equals to 0.025 kg waste/kg renewable feedstock, (source: US-LCI database "Polylactide biopolymer resin at plant kg/RNA"). As assumed in Figure 3, the production efficiency of BB product E_P (how much bio-based feedstock is needed for every unit of BB product) is estimated equal to 1 and no unrecoverable wastes are generated by the process.

In addition, an explorative sensitivity analysis has been performed regarding exclusively the amount of bio-based feedstock content of the BB mulch film, *i.e.* (*i.e.*, $F_{(S)} + F_{(BP)}$), as shown in Figure 4 (Chapter 3).

2.4 Sensitivity analysis

A sensitivity analysis was conducted for BB mulch film to examine the effects of changing the main variables. Given a non-linear dependence of results on parameter values, a Monte Carlo approach (see, *e.g.*, Lloyd and Ries, 2008) has been adopted. The model has been implemented using specifically written routines in the C++ programming language. The model was run with 100,000 events for BB mulch film, where the value of each parameter has been randomly chosen following a Gaussian distribution with a standard deviation within a range of possible and realistic values (Table 5 and **Error! Reference source not found.**; Figure 5 and Figure 6).

3 Results

Considering the characteristics of the films (weight, g/m^2, or thickness, µm, and density, g/cm^3) and the relative functional unit (6000 m^2/ha, Table 3), it is possible to calculate a mass, *M*, that is 90 kg/ha for the BB one. Once calculated the masses, the formulas reported in Table 2 (Chapter 2.2) are applied. Results are shown in Table 4.

Figure 4 shows how the value of the MCI varies according to the percentage variation of the bio-based feedstock in the total mass of the product.

Table 4: *Resulting parameters in the calculation of MCI for BB mulch film.*

Parameter	BB mulch film

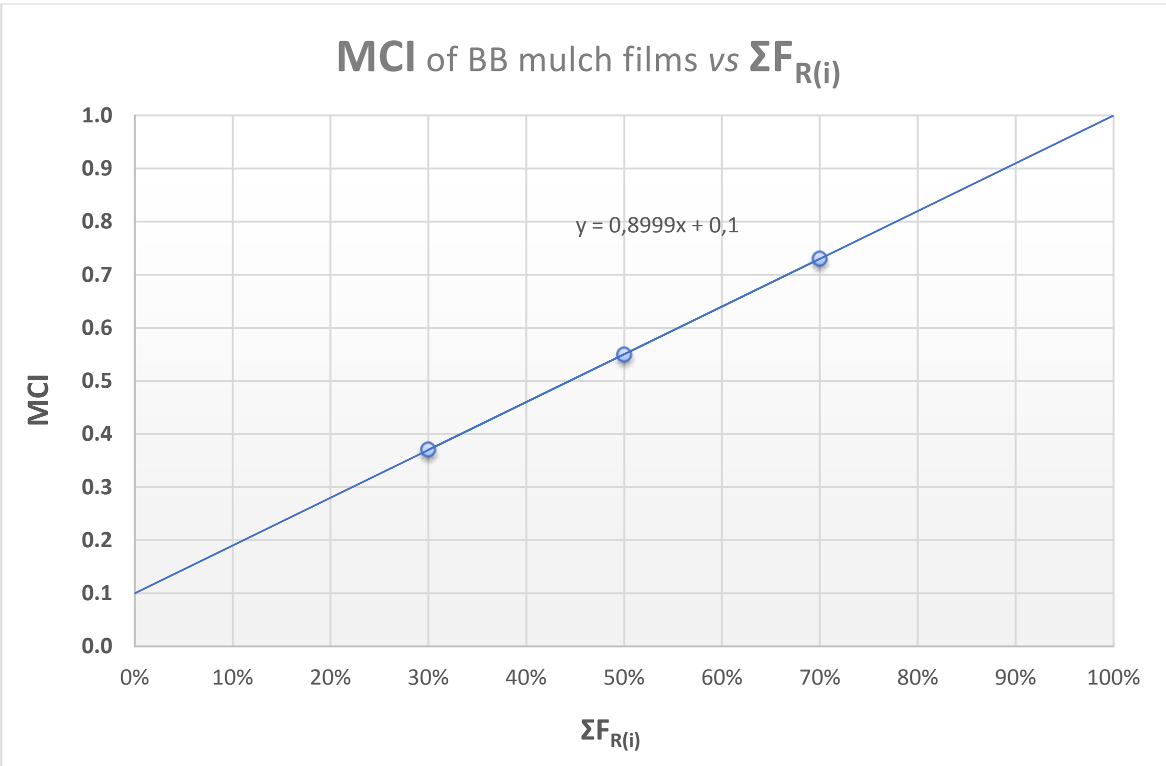

Figure 4: *MCI as a function of* $\Sigma F_{R(i)}$*, the percentage of all the bio-based feedstock/s of the mulch film on mass basis (X-axis).*

3.1 Sensitivity analysis

The results of the sensitivity analysis are presented in the followings Table 5 and Figure 5 and Figure 6. The accuracy band is a fraction of the average and corresponds to a probability of 95%. It has been chosen in order to be representative of the variability of the product category, the BB mulch films. The simulation can thus be regarded as a system composed by a high number of companies, each producing films with different characteristics, that are accounted for in the accuracy band.

Table 5: *Parameters used for the sensitivity analysis of the BB mulch film. (**) The Accuracy Band is defined as twice the standard deviation of the distribution.*

Variable name	**Average**	**Accuracy Band (**)**	**Unit**
M	1000.00	0%	kg
$F_{(S)}/F_{(BP)}$	3.29	10%	fraction
$F_{(S)} + F_{(BP)}$	0.30	30%	fraction
F_U	0.00	0%	fraction
C_U	0.00	0%	fraction
$R_{(S)}$	0.014	100%	fraction
$R_{(BP)}$	0.025	100%	fraction
E_C	1	0%	fraction
E_P	0.95	10%	fraction
C_R	1.00	0%	fraction

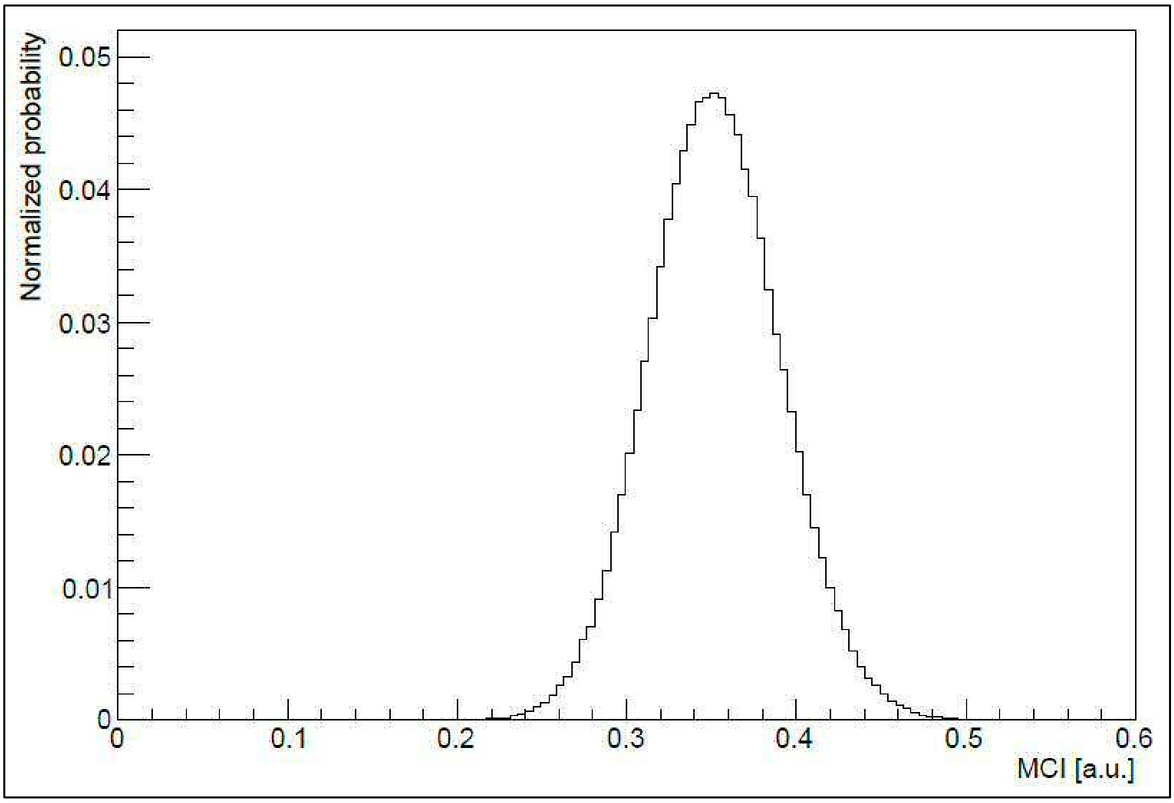

Figure 5: *Resulting distribution of MCI values for BB mulch film.*

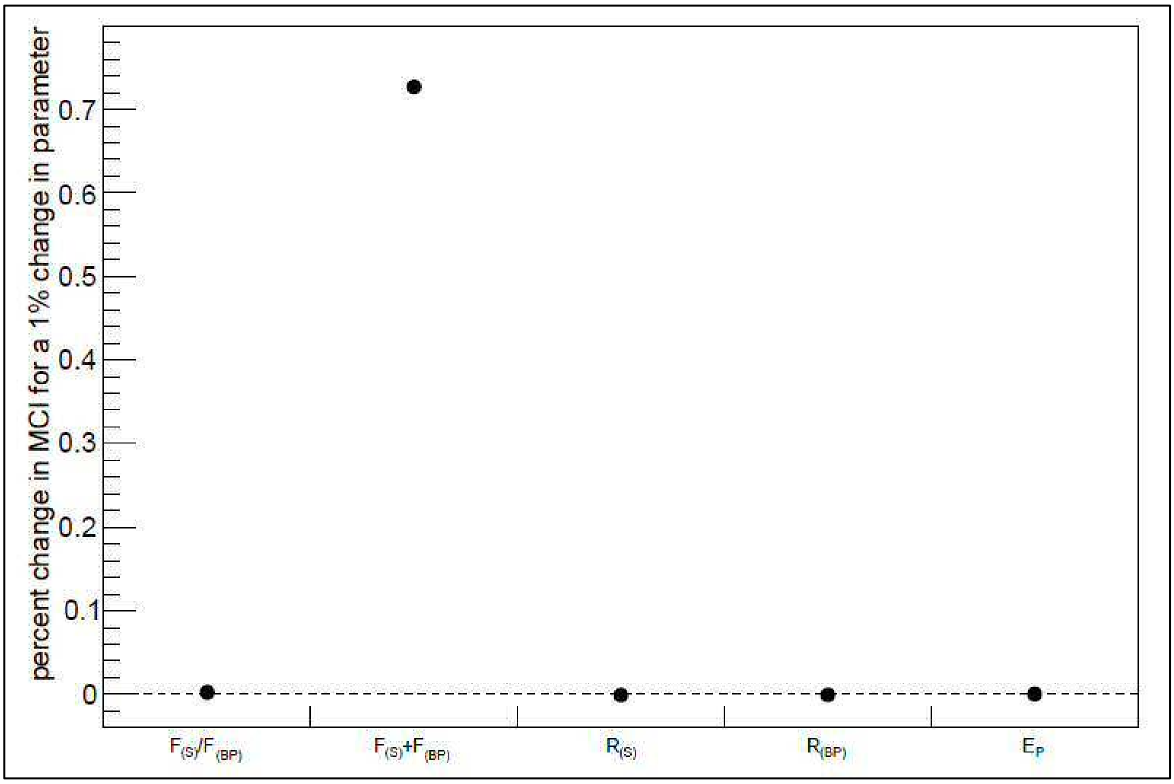

Figure 6: *The most sensitive and relevant parameters in the calculation of the MCI of the BB mulch films.*

4 Discussion

This work applies the principles of the EMF methodology into BB products so as to define common metrics for calculating their circularity. By doing so it proposes some substantial changes to the EMF methodology but still coherent with the overall methodological framework. Such changes should be seen as a generalisation of the methodology provided the following rules are applied:

(1) fossil-based feedstocks or component materials embodied in the BB products whatever is the final disposal (even biological recycling) shall be considered as non-restorative;

(2) bio-based component materials embodied in the BB product that go to biological recycling like composting, or biodegrade in the environment (i.e. BB mulch film) shall be considered restorative as long as they flow through the biosphere safely, without any harm to the environment (e.g. no toxicity effects).

(3) bio-based component materials embodied in the BB product that go to incineration and landfill shall be considered as non-restorative;

The justification of these rules is described in the following.

Fossil-based component materials in the product derive from deposits where they remained stocked for a geological time scale. Once the product is mineralised, its fossil-based portion will be accounted as non-regenerative and therefore linear, due to its origin (Joos et al., 2013). This is true, even if fossil carbon, for example, will re-enter biological cycles, like CO_2 in the atmosphere and other streams, since both fossil-based and bio-based component materials will physically and chemically behave the same, once biodegraded. However, the source of the bio-based carbon was circular before its use (concept of "carbon neutrality", equilibrium between the biogenic carbon released and the carbon absorbed by plants) and will maintain its circularity provided that the carbon is released into the atmosphere at the same rate. The reason has its origin in the EMF general

provisions stating that "biologically sourced materials can only be considered part of a Circular Economy if materials are not used faster than they can be restored naturally" (Ellen MacArthur Foundation & Granta Design, 2015). If BB products are incinerated, the bio-based components are still considered linear, maintaining consistency with EMF principles. Basically, a complete circularity for a BB product is satisfied when its renewable components are 100% bio-based and they go 100% to biological recycling or biodegraded in the environment (for specific application like mulch film).

As for provision (3), a material health rule has its origin in manyfold normative definitions of the CE. In addition, the EMF definition of biological cycles is that of non-toxic materials which are restored into the biosphere and the CE is defined as such if it can "eliminate the use of toxic chemicals". The need of a safety clause has been reviewed under many aspects by Verberne (2016) and can be put as a postulate of the restoration principle: if a flow is toxic it cannot be defined restorative. This is also at the core of the REACH Regulation (EC 1907/2006). In the specific case, the material complies with the standard EN 17033-2018 certifying that no harm is caused to a) all relevant organism groups as plants, invertebrates (e.g. earthworm) and microorganisms, b) important ecological processes maintaining soil functions, c) all relevant exposure pathways as soil pore water, soil pore air and soil material.

A comprehensive approach for MCI calculation should also include non-restorative flows generated at upstream level like biomass growth, in the specific case corn, and biomass conversion processes like starch extraction and refining. Specifically these non-restorative flows correspond to the overall non-recyclable wastes associated to the bio-based feedstock supply thus non-recyclable waste from fertilizer and pesticide production, non-recyclable scraps from conversion processes, etc. In this study such flows of non-restorative waste coming from upstream manufacturing operations were included for the

bio-based feedstocks ($R_{(i)}$) used in manufacturing the BB mulch film applying "cradle to gate" LCA methodology. However, we observed that the inclusion of upstream unrecoverable waste does not significantly influence the MCI results in the chosen case study, since the respective amounts are small. The specific unrecoverable waste for starch and bio-based plasticizer (i.e. kg of waste/kg of bio-based feedstock) were estimated at 0.014 and 0.025, respectively.

The resulting MCI for the 30/70 blend of the BB mulch film is equal to 0.37 in a 0-1 scale and its circularity is linearly linked to the amount of bio-based feedstock used according to the equation $y = 0.89x + 0.1$, where y is the MCI and x is the bio-based feedstock content, therefore the amount of recycled feedstock or (renewable) bio-based feedstock in input is decisive.

Apart from the specific application analysed in this paper, the proposed MCI method can be easily applied and calculated for any kind of BB product as long as the following information are available:

- The bio-based feedstock content, determined according to the standard EN 16785-2:2016, if the composition is known, or directly provided by the BB product manufacturer.
- The End of Life scenario of the studied BB product (real or hypothetical).
- The amount of un-recoverable waste associated to the production of bio-based feedstock contained in the BB product. They can be derived from LCA databases or other specific sources.

5 Conclusions

Bioplastic market is steadily increasing. The value proposition of bio-based and biodegradable products is linked to:

1. the use of renewable feedstock (like starch and its derivates) instead of fossil oil or natural gas;
2. the waste recovery through biological recycling, thanks to their ability to biodegrade in composting facilities or in soil (*e.g.* biodegradable mulch film).

The Material Circularity Indicator (MCI), developed by the EMF, is a metric for quantifying "how much" a product is circular (MCI = 0, fully-linear product; MCI = 1, completely circular product) thus it represents a valuable tool for product eco-design purposes. However, it focuses solely on technical materials, mechanically recycled or reused, leaving out bio-based feedstocks and related biological treatments such as composting. Without common metrics it is not possible to pursue concrete actions, to achieve measurable results and to provide unequivocal references for all products. This research work aims at filling this gap through the development of a methodology coherent with EMF MCI methodology but able to catch the specificities of bio-based and biodegradable products and provide metrics for those innovative products. Direct uses are: (i) supporting the eco-design of innovative bio-based products and (ii) comparing the MCI of BB products with MCI of traditional products (e.g. fossil based).

The proposed method has been applied to a real case study (*i.e.* biodegradable mulch film) providing quantitative metrics about its circularity. Specifically considering a bio-based feedstock content of 30%, the correspondent MCI is 0.37 in a 0-1 scale and its circularity is heavily linked to the bio-based feedstock content according to this relation: $MCI_{(BB\ mulch\ film)} = 0.89 * \text{bio-based feedstock} + 0.1$.

The MCI is a key performance indicator to develop more circular products, in line with the Circular Economy principles. Bioeconomy, thus also BB products, can provide valuable insights in transforming the current (linear) economy in a more circular one, however, the way the biomass is produced, processed and BB products are produced are fundamental aspects to be properly assessed and monitored. This can be done using specific methodologies like LCA. Within this context the proposed MCI has to be seen as a complementary (quantitative) tool for further qualifying the sustainability of BB products.

Declaration of interest

The author declares that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Acknowledgements

The authors thanks prof. Andrea Contin for the fruitful discussion and contribution to the sensitivity analysis, Francesco Degli Innocenti for providing valuable comments and feedback on the topics addressed by the paper and Alessandra Novelli for the general support in the MCI elaboration.

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Dear Reviewers,
The table below provides the requested clarifications and the description of the changes made on the paper for each raised point. Many thanks to both of you for your valuable comments and suggestions. We did our best to improve the paper in the light of the received feedback.

n	Reviewers'comments	Revisions made in the paper
	Reviewer #1	
1	This paper presented a methodological approach for calculating the circularity of bio-based and biodegradable products (mulch films). This research aims at filling this gap through the development of a methodology coherent with EMF MCI methodology but able to catch the specificities of bio-based and biodegradable products and provide metrics for those innovative products. It is a topic of interest to the researchers in the related areas. However, the yield and application range of degradable plastics are important factors affecting their recycling. The whole paper should be reconstructed to make this paper more logically. A major revision is essential before acceptance. The followings are the specific comments.	Many thanks. EU economy has begun taking steps towards a low carbon future (e.g. renewable energy, electric vehicles) and more circular. Bio-based and biodegradable/compostable plastics are seen with interest in all those application where mechanical recycling of traditional plastics is hard to perform. For example in reference to the plastic mulching film the EU market accounts for about 80,000 t/y where >90% is represented by polyethylene (PE) mulch films. The use of PE film < 25 µm is responsible for about 15,000 t/y of microplastics which remain in the soil and about 30,000 t/y of agricultural plastic waste (i.e. PE mulch film) which are dumped or burned in the soil (1). Looking at these figure the great potentialities of developing alternative products results quite evident. However, due to space constraints it is not possible to extensively address these important aspects such as applications of biodegradable plastics, market perspective etc. as suggested by the reviewer. We instead performed some changes in the paper and added two very relevant on-line sources where it is possible to download EU documents, specific reports, case study etc able to direct the reader towards the topics raised by the reviewer. These are: • https://bbia.org.uk/reports/ • https://www.european-bioplastics.org/news/publications/ (1) Revision of the Fertilisers Regulation – benefits of biodegradable mulch films Kristy-Barbara Lange, European Bioplastics, 12 October 2016 http://www.europarl.europa.eu/cmsdata/108931/Kristy%20Barbara%20Lange%20EUBP%20PPT2.pdf
2	Highlights: All of them are exceed the word limits for highlights (less than 85 characters). Please refer to the Guide for Authors.	The highlights have been reduced (see related attach)
3	Table: All tables should be three-line tables in the manuscript.	The tables have been adjusted
4	Line 79 - 82 reference needed	A reference has been added
5	Line 107 Mt, The first appearance should be slightly explained.	The term "Mt" has been expressed as "millions of tonnes"
6	Line 111 - 113 some new references are	New references have been added.

	needed. Please refer to "*Recent advances in toxicological research of nanoplastics in the environment: A review. Environmental Pollution, 2019, 252: 511-521; Microplastic pollution in surface sediments of urban water areas in Changsha, China: Abundance, composition, surface textures. Marine Pollution Bulletin 2018, 136: 414-423.* "	”
7	Line 115 - 117 reference needed	The text has been integrated with the requested time for removing plastic mulch film from the soil and the related reference added
8	Line 137 - 140 Biodegradable polymers are capable of undergoing biological anaerobic or aerobic degradation. A major problem with these plastics is that they have the potential to be biodegraded, but this process requires suitable conditions and microorganisms that are not always reliable in environmental conditions (in situ). The author should explain this point in the article. Please refer to *"Analysis and Prevention of Microplastics Pollution in Water: Current Perspectives and Future Directions. ACS Omega 4(4): 6709-6719".*	The text has been integrated highlighting the importance of the environment’s characteristics on the biodegradation rate of biodegradable bioplastics and the related reference added.
9	Line 284 - 287 reference needed. Although BB mulch films can undergo an ultimate biodegradation with no waste in the soil environment, the biodegradation processes and rate are the keys.	Reference added
10	Line 287 - 291 reference needed.	Reference added
11	Figure 4 should be further revised.	The figure caption has been improved and integrated
12	Line 437 - 438 The authors are encouraged to provide more information and discussion on the eco-design of innovative bio-based products.	The text has been integrated
	Reviewer #2	
13	This manuscript addresses an important topic - how to measure the circularity for a future circular bioeconomy. The suggested approach is novel and it is very good that the approach was demonstrated by the case study of mulch films.	Many thanks
14	It should be recognised that a circularity indicator like MCI is based on material	Absolutely agree. In the paper we only addressed the MCI of bio-based and biodegradable products as additional

	flow analysis only. Thus it does not provide a full picture of sustainability: mass efficiency is not a guarantee of many important sustainability issues like climate change, land use, water use and other resources depletion. This needs to be better elaborated in the paper	metric for further qualifying and assessing bio-based products. This aspect has been further highlighted in the conclusions (line 458-468)
15	The author addressed the toxicity as one of the sustainability aspects but it is: 1) not covered by MCI by definition, and 2) not about life cycle toxicity, which is also an important aspects for biobaesd production (especially in the agricultural phase).	The absence of toxicity is a *sine qua non* condition of the MCI methodology (line 375). It means that if a BB product causes toxicity effects the MCI does not apply since a fundamental principle (i.e. product safety) is not met. Translating this principle into biodegradable mulch film case study we recalled its compliance with the ISO 17033 standard since it encompasses the criteria regarding toxicity aspects beyond other requirements. That said if a BB mulch film is certified according to the ISO 17033 we can consider it safe for the environment.
16	The authors imply to re-define 'waste' (i.e. a material stream that cannot be recovered/biodegraded, or a material stream from a fossil-based source, see lines 285-293). This definition of 'waste' is very different from the definition of EU waste directive. This deviation should be brought into discussion. For example, the authors define that the stream goes to a landfill should be considered not recoverable. Use the case study of BB mulch films - will they biodegrade in a landfill? If yes, why should they be considered waste in this study? This is a very vague line that could practically hinder the application of a new metric.	It is not a re-definition of the term "waste". We have just defined the conditions for judging if a material stream is regenerative or not according to the proposed methodology. MacArthur methodology defines all material streams that go into incinerator or landfill "not regenerative" (i.e. no circular). Similarly we assumed that all BB product streams that go to landfill or incinerator are not regenerative with an exception: the "fossil part" that may constitute a BB product, even if it goes to biological recycling, it is still considered "not regenerative" since its origin is not biogenic. This methodological choice guarantees that a BB products gets a MCI =1 (complete circularity) only if it satisfies at the same time the following conditions: 1) the BB product is 100% made of renewable raw materials and 2) its end of life is represented by 100% biological recycling (composting or AD) or biodegradation in the environment depending on the BB application. Always according to this choice even if a 100% renewable BB product goes to incinerator or landfil thus it emits biogenic CO_2 that goes into the atmosphere and biomass following a circular cycle, this is not considered a regenerative stream since the end of life option does not correspond to that a compostable product has been conceived for (i.e biological recycling). For this reasons MCI will be <1. This is the rationale of the MCI methodology. That said it is not our intention to modify or distort the current definition of "waste".
17	In the case study, the life cycle 'waste' streams from potato/corn/wheat cultivation are not clearly given. The mass balances shown in Figure 3 do not	The Figure 3 has been improved by removing all figures which were not useful for the calculation example. We are sorry for the trouble. In reference to your question about the amounts of agricultural feedstocks they have to be

	added up well: for example, in the case of 30/70 starch blend, the total biomass required is 0.32t corn + 1.26t potato + 0.49t wheat = 2.07t (is this dry mass or green mass?), this gives 0.23t of starch. What is the 2.07-0.23 = 1.84t of the loss? The explanation in line 298 of R(s) of 0.014kg waste per kg renewable feedstock does not seem justified by the numbers in Figure 3.	interpreted as 0.32 kg of corn or 1.26 kg of potato or 0.49 kg of wheat. They are the amounts needed to obtain 0.23 kg of starch (dry matter) which goes into the formulation. All the reported amounts of Figure 3 on starch, plasticizer and polymer refer to dry matter. Now the figure 3 should be clearer. In reference to 0.014 kg of not recoverable wastes per kg of renewable feedstock they refer to the "cradle to gate" LCA boundaries of starch. In the calculation we considered W_F associated to the starch as follows 0.23 * 0.014 = 0.0032 kg/kg BB product.
18	Similar to the comment above: the case study seems completely ignored the the mass loss of the production of fossil-based biodegradable polymer.	In this specific case study the production of BB product (i.e. mulch film) yield is very close to 1 (possible scraps are internally reused in a closed loop), however, the proposed formula for W_F encompasses the mass losses since the process yield is at the denominator of the formula.
19	The effort of a monte carlo simulation is appreciated but is rather over complicated for the conclusion that F(s) + F(BP) is the most sensitive factor - it can be easily derived from a much simpler method like a regular sensitivity analysis.	A global sensitivity analysis can reveal the effect of the co-variation of all parameters, showing how the variance cancels out or add to the specific variation of a factor; the analysis showed to what extent the value of 0.37 can be considered robust, in consideration of all possible variation in defined ranges. The analysis showed that, all possible variations accounted, the standard deviation is 0.041, meaning that 95% of observation would range between 0.29 and 0.45. Not all parameters have a linear effect here. Ep, in particular, as it is placed in the denominator, might have had a relevant effect; its effect here is relatively small and negligible due to its small variation. A sensitivity analysis OAT (one factor at the time) , also known as local sensitivity analysis, or an error propagation would suit this case and indicate which are the most sensitive factors. However, as this paper aims at clarifying the meaning and the robustness of the measure, we opted for a thorough analysis
20	The sensitivity analysis should discuss the influence of the missing data (see comments 4 and 5 above) or input data that are highly uncertain	The uncertainty here is measured when assigning all the factors an accuracy band. R(s) vas assigned a variation of 100% thus largely covering possible changes in the manufacturing process. As for the mass loss see explanation relative to point 4.
21	The discussion section should reflect on the limitation of this new metric.	Conclusions have been improved pointing out that MCI is just a further metric for characterizing BB products.
22	The case study demonstrated a blend material. How would it work for a copolymer which has partially biobased content, such as 30% biobased PET? or partially biobased PBAT (from biobased succinic acid). There should be a clear definition of biobased content (mass), especially for the non-carbon elements	For BB products that contain both biogenic and not biogenic feedstocks, like in the calculation example (Figure 3), only the amount of biogenic feedstock can be considered regenerative. The complementary amount does not. The determination of the regenerative amount thus its complementary not regenerative one is described in the recalled standard EN 16785-2:2016 (line 245)

	such as H, O and even N.	
23	Section 2.4, line 314: justify why a Gaussian distribution is chosen.	All values represent a realisation of industrial processes . The law of large numbers applies here. There is no reason to suspect that a given value would have a different distribution
24	the first para under section 3 Results should be shifted to methodology.	The first para has been moved under methodology section.
25	figure 3: what is the purpose of showing land use and what are the values in cubic meters?	The figure has been adjusted removing the information not needed for the paper purposes. Sorry for the trouble.
26	figure 6: is an illustration needed for the message in the figure?	The figure shows the percent change in the MCI when changing the indicated parameters of + 1%. So, as an example, Fs/F(BP) 3.29 a 1% change (+ 0.03) does not change the MCI; while a change of 1% of Rs (0.014 + 0.0001) yields a change of 0.7% in the MCI
27	- In lines 442-442 in the conclusions section, a relation of MCI of BB mulch films is given. This relation is only based on three data points, which is insufficient to draw a generic conclusion	Actually it is just a graphic representation of the MCI values obtainable through the application of the formulas reported in table 2. The three points represent the three different hypothetical compositions of the BB mulch film (i.e. renewable content equal to 30%, 50% and 70% respectively). For equal end of life (i.e. 100% biodegradation in soil) the MCI increases in function of renewable feedstock content.

Metrics for quantifying the circularity of bioplastics: the case of bio-based and biodegradable mulch films

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Abstract

The concept of circularity and its quantification through the Material Circularity Indicator (MCI) is well established for traditional plastic products. In this paper a methodological approach for calculating the circularity of bio-based and biodegradable (BB) products is proposed and applied to BB mulch films. BB products are different from traditional products in as much as they are sourced and regenerated (recycled) not through technical cycles but the biological loop. The suggested method is an adaptation of the MCI where two major changes were made: (i) the mass of the bio-based component corresponds to the recycled material in input and (ii) the mass of the bio-based component leaving the system through composting or biodegradation in soil is accounted as recycled. The modified MCI supports the Eco-design of innovative BB products and allows for the comparison of their circularity taking into account the biological source and the expected end of life process such as biodegradation. To demonstrate the adaptation, the method has been applied to BB mulch film. Results showed that the MCI of a biodegradable mulch film, characterized by an average bio-based feedstock content of 30% is 0.37 ± 0.04 in a 0-1 scale. For BB mulch film, the amount of bio-based feedstock is the most sensitive factor and controls linearly the value of the MCI.

Keywords: circularity indicators, circular economy, bioplastics, biodegradable mulch film, bio-based product, biodegradation

~~**Abbreviations**~~	
~~**BB**~~	~~Biodegradable and bio-based~~
~~**CE**~~	~~Circular Economy~~
~~**d.m.**~~	~~Dry matter~~
~~**EMF**~~	~~Ellen MacArthur Foundation~~
~~**LCA**~~	~~Life Cycle Assessment~~
~~**LDPE**~~	~~Low-Density Poly-Ethylene~~
~~**MCI**~~	~~Material Circularity Indicator~~
~~**NRF**~~	~~Non-Restorative Flows~~
~~**PBAT**~~	~~Polybutylene adipate terephthalate~~
~~**PE**~~	~~Poly-Ethylene~~
~~**PLA**~~	~~Polylactic acid~~

Abbreviations	
BB	Biodegradable and bio-based
CE	Circular Economy
d.m.	Dry matter
EMF	Ellen MacArthur Foundation
LCA	Life Cycle Assessment
LDPE	Low-Density Poly-Ethylene
MCI	Material Circularity Indicator
NRF	Non-Restorative Flows
PBAT	Polybutylene adipate terephthalate

PE	Poly-Ethylene
PLA	Polylactic acid
PHB	Poly hydroxy butyrate

1 Introduction

To overcome today’s unsustainable model ~~of ‘take~~of ‘take-make-dispose’ and its related risks such as hikes in raw material prices, pressures on the environment, shortage of global resources and waste sinks, a circular approach needs to be applied. It is a new regenerative economic view, based on a balance between economy, environment and society, a total resource efficiency and a Zero Emission Strategy that aims to maximize products value with zero, or minimal, environmental impact (Ghisellini et al., 2016) . Together with structural changes in environmental legislation, new logistics, technologies and sharing schemes, the Circular Economy (CE) approach which is regenerative by design, aims at closing materials loops, *i.e.* at reducing virgin materials input and waste output.

In December 2015, the European Commission developed an Action Plan for Circular Economy (European Commission, 2015), where plastic was considered a priority to be tackled. In January 2018, an *EU Plastic Strategy* (European Commission, 2018) was adopted, in order to react to the increasing environmental problems concerning plastic production, consumption, use and disposal along the same lines of the CE approach. Two fundamental steps to increase the circularity of different plastic products are (i) the abandonment of fossil fuels, *i.e.* currently 90% of the plastic is produced by virgin petroleum-based feedstock (Ellen MacArthur Foundation, 2017), and (ii) the development of easily recyclable products which are recycled. Today, in EU the share of plastics collected for recycling is 30% while the use of recycled plastics is just 6% (European Commission, 2018).

Biodegradable and bio-based (BB) plastics are spreading across markets (Institute for Bioplastics and Biocomposites, 2018) as a valid contribution to meet CE aims and principles. This is true as long as the supply of renewable raw materials, generally from agriculture, is based on a sustainable approach and the conversion processes along the

supply chain are efficient and highly integrated in a Life Cycle Assessment (LCA) perspective (EPLCA – European Platform on LCA). While traditional plastics can be mechanically recycled or incinerated with energy recovery, BB plastic products offer new recycling routes in waste management, due to their biodegradability. Organic recycling (through composting or anaerobic digestion) or in the case of specific applications such as agricultural mulch films, biodegradation in the environment, offer additional recovery options resulting in less wastes and less contamination of soil by plastic residues (Razza et al., 2012; Lange, B., 2016). An extensive literature review about the potentialities and benefits of renewable and compostable bioplastics, encompassing market perspective, applications, economic effects etc. can be found here: (BBIA; European Bioplastics).

Nevertheless, the research and development of innovative products, such as the BB products, implies the development of methodologies and metrics capable of measuring their circularity. Without this it is not possible to achieve measurable results and improving actions, as well as provide unequivocal references for comparisons of products of the same type/category. In 2015 the Material Circularity Indicator (MCI) was developed (Ellen MacArthur Foundation & Granta Design, 2015) which aims to quantify the regeneration of a product's material flow and is considered one of the few, among sixteen CE indexes suiting a micro-scale assessment of circularity at product or company level (Lonca et al., 2018). However, it focuses solely on technical cycles and recycled materials. Furthermore, recovery and recycling through the biological cycle offered by industrial composting, anaerobic digestion or biodegradation in natural environments are not considered as end of life options. In order to apply the MCI system to BB plastic products, the development of an enhanced methodology is necessary.

The approach proposed by the authors allows to quantify the circularity of BB plastic products ~~(*e.g.* starch-based bioplastics)~~ and to make comparisons with equivalent traditional plastic products. To demonstrate the applicability of the proposed method a computational example for mulch film products is provided. In so doing so, the paper aims at contributing to the Eco-design of these innovative products.

1.1 The case study of mulch films

Plastic mulch films represent an important agronomical technique well established for the production of many crops thanks to numerous agronomical advantages such as: increased yield and higher quality of productions (Steinmetz et al., 2016) ; weed control and reduced use of pesticides; early crop production and reduced soil moisture loss (Briassoulis and Giannoulis, 2018). As a consequence, the plastic films consumption has increased year-by-year, reaching a current global market estimated at 1.4 millions of tonnes ~~Mt~~, mainly in Asia (Briassoulis and Giannoulis, 2018; Mormile et al., 2017) , and covering 80,000 km^2 of agricultural surface (0.6% of the global arable land). The mulch film market in Europe is estimated by Agriculture Plastic & Environment and by the European Bioplastic Associations at 76-80 kt. The most used raw material is Poly-Ethylene (PE) in its different forms, due to its processability, chemical resistance, high durability and flexibility (Kasirajan and Ngouajio, 2012; Plasticulture, 2016 and 2018; Shen, M. et al., 2019; Wen, X. et al., 2018).

Despite these benefits, manifold environmental and agronomic problems have been pointed out. After its useful life – which in general does not exceed 1 to 3 months – the mulch film has to be removed and properly disposed of, a time-consuming (about 16 hours per hectare) and costly procedure (Scaringelli, M., 2016; Briassoulis, D., 2013). The recovered film is usually heavily contaminated with soil and organic residues, making mechanical recycling technically difficult and not a cost-efficient solution (Briassoulis et al., 2018; Figuier, 2016; De Lèpinau, Arbenz, 2016). The most common end of life of collected films in Europe is still landfilling (about 50%), followed by energy recovering

and finally mechanical recycling (Le Moine, 2014). Recent Chinese prohibition (January 2018) to import different types of wastes is heavily impacting the European agricultural plastic waste management, highlighting the difficulty in properly recycling this type of plastics (Tamma, 2018). Plastic films may not be properly collected and recycled but disposed of by burning in the field or by uncontrolled landfilling or left directly in the (agricultural) soils, causing serious environmental concerns. An example is the “White pollution” phenomena described in the Xinjiang Autonomous Region (China), in which the residual plastic film can reach 200 kg/ha in the top soil with detrimental effects on soils’ quality, health and fertility (Liu, He, & Yan, 2014; Gao *et al.*, 2019; Steinmetz *et al.*, 2016).

As a reaction, there has been significant research into novel materials especially related to biodegradable and bio-based (BB) mulch films, which enable an effective biodegradation in soil and provide comparable agronomical performances (Touchaleaume et al., 2016). The term “bio-mulch film” brings together several types of both bio-based and fossil oil-based biodegradable polymers and blends of them, such as polylactic acid (PLA), polybutylene adipate co-terephthalate (PBAT), starch-based polymer blends or copolymers. They biodegrade when exposed to bioactive environments such as soil and compost (Kasirajan et al., 2012) which means that they can be left *in situ* to be fully biodegraded after being used. Clearly the biodegradation rate of biodegradable bioplastics is influenced by the environmental conditions such as the types of available bacteria, fungi thus specific enzymes namely native microflora (Pico, Y. et al., 2019). However their intrinsic biodegradability ~~must be proved by accredited certification bodies and standardized procedures~~ allow the complete biodegradation with times similar to natural polymers such as cellulose used as reference by the relevant standards and certification schemes.

The EN 17033:2018 is a new European Norm (standard) concerning “Plastics - Biodegradable mulch films for use in agriculture and horticulture - Requirements and test methods”, which sets the necessary tests and limits to define biodegradability, performances and environmental impacts of BB much films. The material is considered completely biodegradable if it achieves a complete biodegradation (absolute or relative to the reference material) in a test period no longer than 24 months (mineralization into CO_2). Additionally, a control of constituents (such as metals) and eco-toxicity testing (acute and chronic toxicity tests on plant growth, earthworm; nitrification inhibition test with soil microorganisms) were required. A certified mulch film guarantees that the product will completely biodegrade in the soil without adversely impacting on the environment.

1.2 Goal of the paper

The goal of the paper is to provide a general and common metric to measure the circularity of a bio-based and biodegradable (BB) product and to apply the methodology at product level to a category of products, namely bio-based and biodegradable mulch films.

2 Materials and Methods

2.1 MCI accounting according to the EMF methodology

The Material Circularity Indicator (MCI), according to the Ellen MacArthur Foundation (EMF) methodology (Ellen MacArthur Foundation & Granta Design, 2015), is a number that can range from 0 (pure linearity) to 1 (pure circularity). A purely linear production provides for the exclusive use of virgin raw materials that turn into waste at the end of the use phase of the product. Vice-versa, pure circularity includes the use of recycled materials and does not produce wastes (regenerative streams). Circularity can be achieved in different ways: as for the purpose of this paper, only recycling will be considered since

reuse is not an option for thin biodegradable mulch films. Since the method considers only mass flows, the recycling corresponds to the recovery of materials for the original purpose or for other purposes and excludes energy recovery, considered as a loss of materials equal to landfill disposal. The materials recovered feed back into the process as recycled feedstock.

The MCI methodology differentiates ‘technical cycles’ from ‘biological cycles’, modelling only the former. The first contains products and materials re-entering into the system (market) with the highest possible qualities and for as long as possible (thanks to reuse, repair, refurbishment and recycling) and the latter includes biological materials used in cascade until their restoration into the biosphere and the re-constitution of natural resources.

The material flows associated to the production of a generic technical cycle from non-renewable sources are summarized in Figure 1~~Figure 1~~. The dashed lines indicate that recycled feedstock does not have to be sourced from the same product but can be acquired on the market. With reference to Figure 1~~Figure 1~~, the list of the parameters used in the EMF methodology is reported in Table 1~~Table 1~~, while the equations relevant for the analysis carried out in this paper are described in the following sections (Table 2~~Table 2~~, Chapter 2.2).

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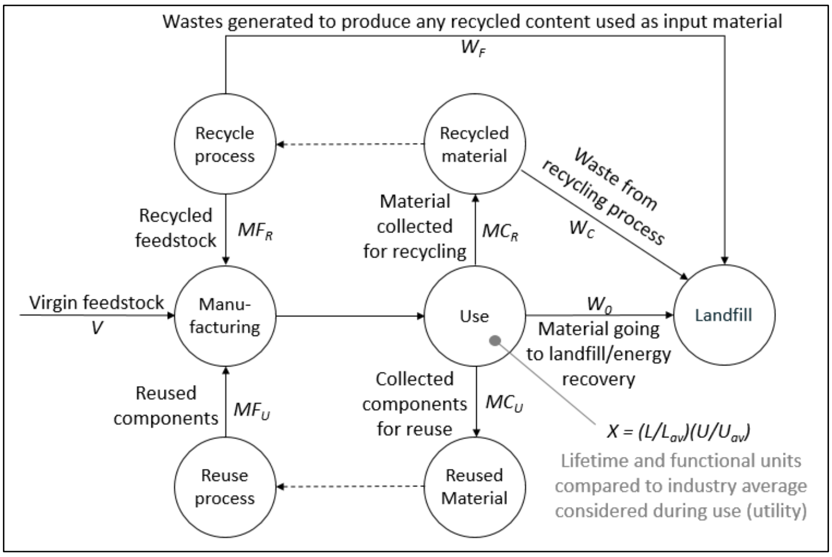

Figure 1: *Diagram of material flows and associated variables of a generic product (modified from Ellen MacArthur Foundation & Granta Design, 2015).*

Table 1: *Parameters and relative definitions used in the EMF methodology.*

~~**Parameter**~~	~~**Definition**~~
~~M~~	~~Total mass of the product~~
~~F_R~~	~~Fraction of mass of a product's feedstock from recycled sources~~
~~F_U~~	~~Fraction of mass of a product's feedstock from reused sources~~
~~V~~	~~Mass of virgin feedstock used in a product~~
~~C_R~~	~~Fraction of mass of a product being collected to go into a recycling process~~

C_U	Fraction of mass of a product going into component reuse
E_C	Efficiency of the recycling process used for the portion collected for recycling
E_F	Efficiency of the recycling process used to produce recycled feedstock for a product
W	Total mass of unrecoverable waste associated with a product
W_0	Mass of unrecoverable waste (landfill, waste to energy and any other type of process where the materials are no longer recoverable)
W_C	Mass of unrecoverable waste generated in the process of recycling parts of a product (after use)
W_F	Mass of unrecoverable waste generated when producing recycled feedstock for a product
X	Utility of a product, calculated as $X = (L/L_{av})(U/U_{av})$
L	Actual average lifetime of a product
L_{av}	Actual average lifetime of an industry-average product of the same type
U	Actual average number of functional units achieved during the use phase of a product
U_{av}	Actual average number of functional units achieved during the use phase of an industry-average product of the same type

Parameter	Definition
M	Total mass of the product
F_R	Fraction of mass of a product's feedstock from recycled sources
F_U	Fraction of mass of a product's feedstock from reused sources
V	Mass of virgin feedstock used in a product
C_R	Fraction of mass of a product being collected to go into a recycling process
C_U	Fraction of mass of a product going into component reuse
E_C	Efficiency of the recycling process used for the portion collected for recycling
E_F	Efficiency of the recycling process used to produce recycled feedstock for a product
W	Total mass of unrecoverable waste associated with a product
W_0	Mass of unrecoverable waste (landfill, waste to energy and any other type of process where the materials are no longer recoverable)
W_C	Mass of unrecoverable waste generated in the process of recycling parts of a product (after use)
W_F	Mass of unrecoverable waste generated when producing recycled feedstock for a product
X	Utility of a product, calculated as $X = (L/L_{av})(U/U_{av})$
L	Actual average lifetime of a product
L_{av}	Actual average lifetime of an industry-average product of the same type
U	Actual average number of functional units achieved during the use phase of a product
U_{av}	Actual average number of functional units achieved during the use phase of an industry-average product of the same type

The Material Circularity Indicator is determined as follows: ,
where *LFI* is the Linear Flow Index measuring the flows of virgin materials and unrecoverable wastes associated to the examined product.

A function of the utility, –, is used to correct the *LFI*. The function *F* is chosen in such a way that improvements of the utility of a product (e.g., by using it longer) have the same impact on its MCI as a reuse of components, leading to the same amount of reduction of virgin material use and unrecoverable waste. Setting a = 0.9, MCI takes, by convention, the value 0.1 for a fully linear product (*i.e.*, *LFI* = 1) whose utility equals the industry average (*i.e.*, *X* = 1). This leaves some margin to distinguish between processes with a high linearity but different utilities.

2.2 MCI accounting for bio-based and biodegradable (BB) products

To apply the EMF methodology to BB products, formulas and flows (Figure 1~~Figure 1~~ and Figure 2~~Figure 2~~) are adapted as it follows:

1. The fraction of the recycled feedstock, F_R, corresponds to the share of the bio-based feedstock content in the final BB product, $F_{R(i)}$. It is the ratio of the d.m. amount of bio-based feedstock per d.m. amount of the total mass of BB product (EN 16785-2:2016).
2. The fraction of restorative mass going into a recycling process, C_R, corresponds to the share of bio-based feedstock content in the BB product biologically recovered (*e.g.* through composting) or biodegraded in the natural environment, as it happens for specific applications (e.g. biodegradable mulch film, *etc.*). It is the ratio of the d.m. amount of bio-based feedstock per d.m. amount of the total mass of BB product that is biologically recycled.

The modified scheme is shown in Figure 2~~Figure 2~~. Table 2~~Table 2~~ lists the formulas as adapted to BB products.

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Table 2: *List of formulas as developed by EMF methodology compared to the proposed adaptation to BB products.*

~~EMF methodology~~	~~Adaptation to BB products~~
EMF methodology	Adaptation to BB products

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The mass of fossil-based feedstock which may be contained in BB products (V) is obtained as a difference of the total mass (M) minus the bio-based fraction; in this case the F_R in the EMF methodology corresponds to the sum of the fractions of all the bio-based feedstock/s used in manufacturing the BB product. Therefore, is the total bio-based feedstock mass in the product. In single-use products, such as mulch films, reuse is not considered for BB products, so that $F_U = C_U = 0$.

W_F is the total amount of unrecoverable waste associated to the production of bio-based feedstock used to produce BB products (*i.e.* the amount of uncoverable waste per unit of BB product). Bio-based feedstocks such as starch, ~~and~~ PLA, PHB etc. generate non-restorative flows which can be quantified. Such unrecoverable waste correspond to $R_{(i)}$, the specific amount of waste generated within cradle-to-gate boundaries per unit of bio-based feedstock going into manufacturing, and it is estimated through LCA studies. Thus all inputs from growth and harvesting phases and the related wastes generated by fertilisers and pesticides are here accounted. $R_{(i)}$ can be easily found in specific literature or life cycle inventories (LCI) present in LCA databases. In the calculation of W_F, also the efficiency of manufacturing process of BB products E_P is considered, as the ratio of the overall bio-based feedstock content in the final BB product to the bio-based feedstock in input to the manufacturing process.

The material flows associated to the production of a generic BB product are summarized in Figure 2~~Figure 2~~.

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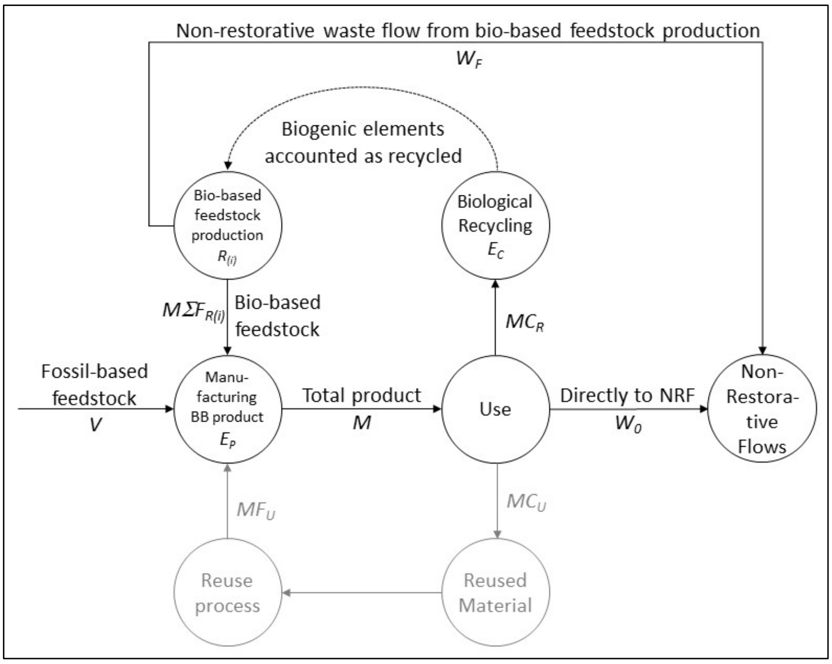

Figure 2: *Description of material flows adaptation to BB products; in this paper, the reuse flow is out of scope ($C_U = F_U = 0$).*

The biodegradation of bio-based feedstock does not imply the generation of waste W_C as it occurs in a standard mechanical recycling process. This implies that C_R and E_C (*i.e.* the efficiency of the biodegradation process) equal to 1. Indeed, a BB raw material, sent to biological treatment (composting) or biodegraded in a natural environment, is fully transformed in its chemical elements (C, H and O mainly) derived from the decomposition of complex molecules (polymers) without the release of waste (Witt et al., 2001; Marten et al., 2003; Eubeler et al., 2010; BASF, 2018; Institute of Bioplastics and Biocomposites, 2018; OWS, 2018; Zumstein et al., 2018). These natural elements return into the environment and are then available in the respective biogeochemical cycles. The (biodegradable) fossil portion behaves as well; consequently, $W_C = 0$.

Nevertheless, the fossil-based feedstock cannot be considered as a regenerative circular feedstock, since it derives from carbon stored for millions of years and extracted by man, not being part of the active and fast biogeochemical carbon cycle. This is accounted in the quantification of W_0, the mass of unrecoverable waste from use (*i.e.* the linear stream going to landfill or incineration, the Non-Restorative Flows, NRF), as , the total amount of fossil-based feedstock.

Since W_F and W_C are associated to complete different processes and W_C is always equal zero, the double counting issue does not occur and the quantification of W and *LFI* is modified as reported in Table 2~~Table 2~~.

2.3 MCI calculation for mulch films: scope, inventory and assumptions

The new formulas reported in Table 2~~Table 2~~ were applied to a single use product namely a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1.

In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the BB film has been arbitrarily ~~is~~ assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e. 23% of starch, $F_{(S)}$, and 7% of a bio-based ~~plasticizer~~additive, $F_{(BA)}$), while the rest was assumed to consist of fossil feedstock (

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Figure 3Figure 3). Since a generalized approach was used and no primary data were implemented, the information were extrapolated from literature (Institute of Bioplastics and Biocomposites, 2018)–; the main characteristics of the two examined products are presented in Table 3Table 3.

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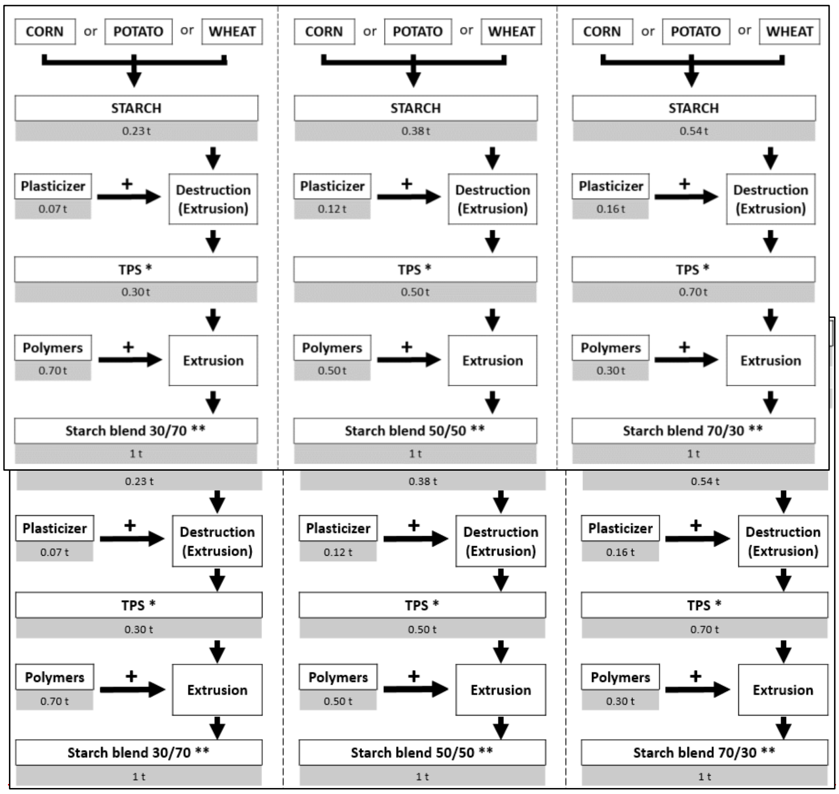

Figure 3: *Examples of hypothetical ~~starch~~bio-based polymers; in this paper, the first option on the left (starch blend 30/70) has been chosen ~~as representative of a BB mulch film.~~for carrying out the numerical MCI calculation (working hypothesis). The figure considers a 100%-efficiency in every phase of production, so that the residues are equal to zero; the same assumption is done in this paper. *TPS (Thermoplastic starch), starch content 75%; **Ratio TPS/Polymer; modified from Institute of Bioplastics and Biocomposites, 2018.*

Table 3*: Key features representative of the BB mulch films.*

	~~**BB mulch film**~~
~~**Material**~~	~~30% bio-based feedstock (23% starch + 7% bio-based plasticizer) + 70% fossil-based feedstock~~
~~**Thickness** (µm)~~	~~12~~
~~**Density** (g/cm³)~~	~~1.25~~
~~**Weight** (g/m²)~~	~~15.2~~
~~**Functional unit** (the covering of the agricultural land)~~	~~6000 m²/ha (the actual mulched soil in a hectare is generally equal to the 60% of the total area; Malinconico, 2017)~~

	BB mulch film
Material	30% bio-based feedstock (23% starch + 7% bio-based additive) + 70% fossil feedstock
Thickness (µm)	12
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Weight (g/m²)	15.2
Functional unit (the covering of the agricultural land)	6000 m²/ha (the actual mulched soil in a hectare is generally equal to the 60% of the total area; Malinconico, 2017)

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In the calculation of MCI for the BB mulch film, the adapted formulas were used together with assumptions. As stated before, BB mulch films are blends of bio-based and fossil based feedstocks (in the specific case, 30% and 70% respectively). Unlike the LDPE mulch film that has to be removed and disposed of, the BB mulch film is left in soil where it undergoes an ultimate biodegradation (so that $C_R = 1$) with no waste (so that $E_C = 1$), in respect of the specific standard EN 17033:2018. As a result of polymers' decomposition, the derived (biogenic) C, H and O finally return into biosphere (atmosphere, microorganism biomass, organic material pool) (OWS, 2018), and back into biogeochemical cycles in a relatively short time ("Biogenic elements accounted as recycled" in Figure 2Figure 2), with the exception of -humified compounds. Actually, also C, H and O deriving from fossil-based sources undergo biodegradation (Zumstein, M.T., 2018) but they are not considered as a regenerative flow ("Waste from non-restorative flow" in *Figure 2Figure 2*) and their "wastes" are indeed calculated in W_0.

Applying a conservative approach, W_F, the waste generated by the production of each bio-based feedstock, is quantified considering a "cradle to gate" LCA study. The estimated solid wastes $R_{(i)}$ for the presented case study are related to the production of starch ($F_{(S)}$), with an amount $R_{(S)}$ of 0.014 kg of waste per kg of renewable feedstock (source: personal communication A. Novelli), and to the production of the bio-based additive plasticizer ($F_{(BAP)}$), with $R_{(BPA)}$ equals to 0.025 kg waste/kg renewable feedstock (US-LCI database), (source: US-LCI database "Polylactide biopolymer resin at plant kg/RNA"). As assumed in

Figure 3Figure 3, the production efficiency of BB product E_P (how much bio-based feedstock is needed for every unit of BB product) is estimated equal to 1 and no unrecoverable wastes are generated by the process.

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In addition, an explorative sensitivity analysis has been performed regarding exclusively the amount of bio-based feedstock content of the BB mulch film, *i.e.* (*i.e.*, $F_{(S)}$ + $F_{(BPA)}$), as shown in Figure 4~~Figure 4~~ (Chapter 3). Considering the characteristics of the films (weight, g/m^2, or thickness, μm, and density, g/cm^3) and the relative functional unit (6000 m^2/ha, Table 3~~Table 3~~), it is possible to calculate a mass, *M*, that is 90 kg/ha for the BB one. Once calculated the masses, the formulas reported in Table 2~~Table 2~~ (Chapter 2.2) are applied. Results are shown in Table 4~~Table 4~~.

2.4 *Sensitivity analysis*

A sensitivity analysis was conducted for BB mulch film to examine the effects of changing the main variables. Given a non-linear dependence of results on parameter values, a Monte Carlo approach (see, *e.g.*, Lloyd and Ries, 2008) has been adopted. The model has been implemented using specifically written routines in the C++ programming language. The model was run with 100,000 events for BB mulch film, where the value of each parameter has been randomly chosen following a Gaussian distribution with a standard deviation within a range of possible and realistic values (Table 5~~Table 5~~ and **Error! Reference source not found.**~~Table 6~~; Figure 5~~Figure 5~~ and Figure 6~~Figure 6~~).

3 Results

~~Considering the characteristics of the films (weight, g/m^2, or thickness, μm, and density, g/cm^3) and the relative functional unit (6000 m^2/ha, Table 3), it is possible to calculate a mass, *M*, that is 90 kg/ha for the BB one. Once calculated the masses, the formulas reported in Table 2 (Chapter 2.2) are applied. Results are shown in Table 4.~~

Figure 4~~Figure 4~~ shows how the value of the MCI varies according to the percentage variation of the bio-based feedstock in the total mass of the product.

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Table 4: *Resulting parameters in the calculation of MCI for BB mulch film.*

~~Parameter~~	~~BB mulch film~~
Parameter	**BB mulch film**

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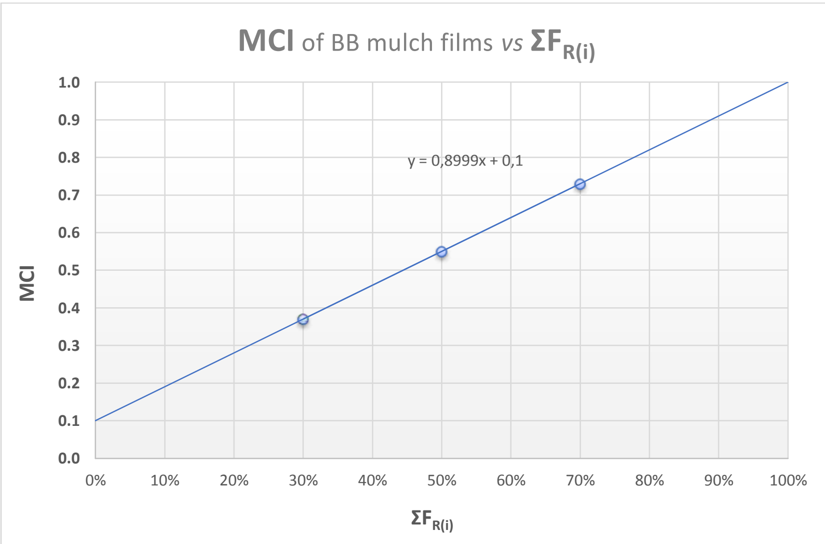

Figure 4: *MCI as a function of the amount of bio-based feedstock/s in the BB mulch film $\Sigma F_{R(i)}$, expressed as ~~$\Sigma F_{R(i)}$,~~ the percentage of all the bio-based feedstock/s of the mulch film on dry mass basis (X-axis). The dots correspond to the three different hypothetical bioplastic compositions of Figure 3.*

3.1 Sensitivity analysis

The results of the sensitivity analysis are presented in the followings Table 5~~Table 5~~ and Figure 5~~Figure 5~~ and Figure 6~~Figure 6~~. The accuracy band is a fraction of the average and corresponds to a probability of 95%. It has been chosen in order to be representative of the variability of the product category, the BB mulch films. The simulation can thus be

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regarded as a system composed by a high number of companies, each producing films with different characteristics, that are accounted for in the accuracy band.

Table 5: *Parameters used for the sensitivity analysis of the BB mulch film. (**) The Accuracy Band is defined as twice the standard deviation of the distribution.*

~~Variable name~~	~~Average~~	~~Accuracy Band (**)~~	~~Unit~~
~~M~~	~~1000.00~~	~~0%~~	~~kg~~
~~$F_{(S)}/F_{(BP)}$~~	~~3.29~~	~~10%~~	~~fraction~~
~~$F_{(S)} + F_{(BP)}$~~	~~0.30~~	~~30%~~	~~fraction~~
~~F_U~~	~~0.00~~	~~0%~~	~~fraction~~
~~C_U~~	~~0.00~~	~~0%~~	~~fraction~~
~~$R_{(S)}$~~	~~0.014~~	~~100%~~	~~fraction~~
~~$R_{(BP)}$~~	~~0.025~~	~~100%~~	~~fraction~~
~~E_C~~	~~1~~	~~0%~~	~~fraction~~
~~E_P~~	~~0.95~~	~~10%~~	~~fraction~~
~~C_R~~	~~1.00~~	~~0%~~	~~fraction~~

Variable name	**Average**	**Accuracy Band (**)**	**Unit**
M	1000.00	0%	kg
$F_{(S)}/F_{(BPA)}$	3.29	10%	fraction
$F_{(S)} + F_{(BPA)}$	0.30	30%	fraction
F_U	0.00	0%	fraction
C_U	0.00	0%	fraction
$R_{(S)}$	0.014	100%	fraction
$R_{(BAP)}$	0.025	100%	fraction
E_C	1	0%	fraction
E_P	0.95	10%	fraction

C_R	1.00	0%	fraction

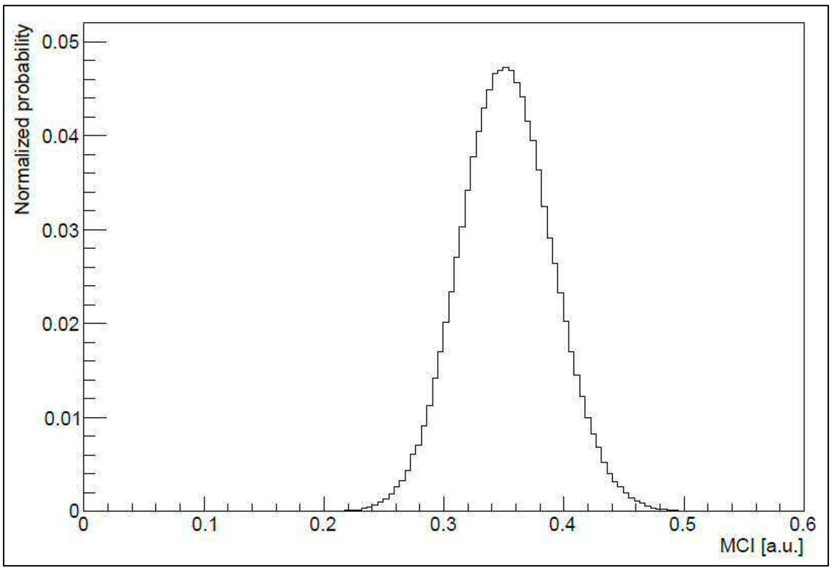

Figure 5: *Resulting distribution of MCI values for BB mulch film.*

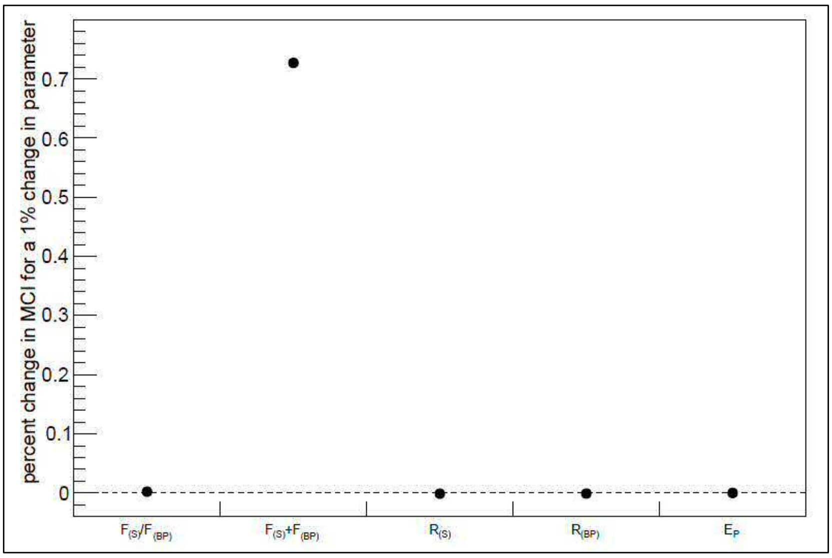

Figure 6: *The most sensitive and relevant parameters in the calculation of the MCI of the BB mulch films.*

4 Discussion

This work applies the principles of the EMF methodology into BB products so as to define common metrics for calculating their circularity. By doing so it proposes some substantial changes to the EMF methodology but still coherent with the overall methodological framework. Such changes should be seen as a generalisation of the methodology provided the following rules are applied:

(1) fossil-based feedstocks or component materials embodied in the BB products whatever is the final disposal (even biological recycling) shall be considered as non-restorative;

(2) bio-based component materials embodied in the BB product that go to biological recycling like composting, or biodegrade in the environment (i.e. BB mulch film) shall be

considered restorative as long as they flow through the biosphere safely, without any harm to the environment (e.g. no toxicity effects).

(3) bio-based component materials embodied in the BB product that go to incineration and landfill shall be considered as non-restorative;

The justification of these rules is described in the following.

Fossil-based component materials in the product derive from deposits where they remained stocked for a geological time scale. Once the product is mineralised, its fossil-based portion will be accounted as non-regenerative and therefore linear, due to its origin (Joos et al., 2013). This is true, even if fossil carbon, for example, will re-enter biological cycles, like CO_2 in the atmosphere and other streams, since both fossil-based and bio-based component materials will physically and chemically behave the same, once biodegraded. However, the source of the bio-based carbon was circular before its use (concept of "carbon neutrality", equilibrium between the biogenic carbon released and the carbon absorbed by plants) and will maintain its circularity provided that the carbon is released into the atmosphere at the same rate. The reason has its origin in the EMF general provisions stating that "biologically sourced materials can only be considered part of a Circular Economy if materials are not used faster than they can be restored naturally" (Ellen MacArthur Foundation & Granta Design, 2015). If BB products are incinerated, the bio-based components are still considered linear, maintaining consistency with EMF principles. Basically, a complete circularity for a BB product is satisfied when its renewable components are 100% bio-based and they go 100% to biological recycling or biodegraded in the environment (for specific application like mulch film).

As for provision (3),– a material health rule has its origin in manyfold normative definitions of the CE. In addition, the EMF definition of biological cycles is that of non-toxic materials which are restored into the biosphere and the CE is defined as such if it can "eliminate the use of toxic chemicals". The need of a safety clause has been reviewed under many aspects by Verberne (2016) and can be put as a postulate of the restoration principle: if a flow is toxic it cannot be defined restorative. This is also at the core of the REACH Regulation (EC 1907/2006). In the specific case, the material complies with the standard EN 17033-2018 certifying that no harm is caused to a) all relevant organism groups as plants, invertebrates (e.g. earthworm) and microorganisms, b) important ecological processes maintaining soil functions, c) all relevant exposure pathways as soil pore water, soil pore air and soil material.

A comprehensive approach for MCI calculation should also include non-restorative flows generated at upstream level like biomass growth, in the specific case corn, and biomass conversion processes like starch extraction and refining. Specifically these non-restorative flows correspond to the overall non-recyclable wastes associated to the bio-based feedstock supply thus non-recyclable waste from fertilizer and pesticide production, non-recyclable scraps from conversion processes, etc. In this study such flows of non-restorative waste coming from upstream manufacturing operations were included for the bio-based feedstocks ($R_{(i)}$) used in manufacturing the BB mulch film applying "cradle to gate" LCA methodology. However, we observed that the inclusion of upstream unrecoverable waste does not significantly influence the MCI results in the chosen case study, since the respective amounts are small. The specific unrecoverable waste for starch and bio-based additive ~~plasticizer~~ (i.e. kg of waste/kg of bio-based feedstock) were estimated at 0.014 and 0.025, respectively.

The resulting MCI for the 30/70 blend of the BB mulch film is equal to 0.37 in a 0-1 scale and its circularity is linearly linked to the amount of bio-based feedstock used according to the equation $y = 0.89x + 0.1$, where y is the MCI and x is the bio-based feedstock content, therefore the amount of recycled feedstock or (renewable) bio-based feedstock in input is decisive.

Apart from the specific application analysed in this paper, the proposed MCI method can be easily applied and calculated for any kind of BB product as long as the following information are available:

- The bio-based feedstock content, determined according to the standard EN 16785-2:2016, if the composition is known, or directly provided by the BB product manufacturer.
- The End of Life scenario of the studied BB product (real or hypothetical).
- The amount of un-recoverable waste associated to the production of bio-based feedstock contained in the BB product. They can be derived from LCA databases or other specific sources.

5 Conclusions

Bioplastic market is steadily increasing. The value proposition of bio-based and biodegradable products is linked to:

1. the use of renewable feedstock (like starch and its derivates) instead of fossil oil or natural gas;
2. the waste recovery through biological recycling, thanks to their ability to biodegrade in composting facilities or in soil (*e.g.* biodegradable mulch film).

The Material Circularity Indicator (MCI), developed by the EMF, is a metric for quantifying “how much” a product is circular (MCI = 0, fully-linear product; MCI = 1, completely circular product) thus it represents a valuable tool for product eco-design purposes. However, it focuses solely on technical materials, mechanically recycled or reused, leaving out bio-based feedstocks and related biological treatments such as composting. Without common metrics it is not possible to pursue concrete actions, to achieve measurable results and to provide unequivocal references for all products. This research work aims at filling this gap through the development of a methodology coherent with EMF MCI methodology but able to catch the specificities of bio-based and biodegradable products and provide metrics for those innovative products. Direct uses are: (i) supporting the eco-design of innovative bio-based products and (ii) comparing the MCI of BB products with MCI of traditional products (e.g. fossil based).

The proposed method has been applied to a real case study (*i.e.* biodegradable mulch film) providing quantitative metrics about its circularity. Specifically considering a bio-based feedstock content of 30%, the correspondent MCI is 0.37 in a 0-1 scale and its circularity is heavily linked to the bio-based feedstock content according to this relation: $MCI_{(BB\ mulch\ film)} = 0.89*bio\text{-}based\ feedstock + 0.1$.

The MCI is a key performance indicator to develop more circular products, in line with the Circular Economy principles like the use of renewable materials and the reduction of the amount of not recoverable waste. MCI will support the development of innovative products just based on these two important characteristics specific for each BB product/application and end of life scenario. Bioeconomy, thus also BB products, can provide valuable insights in transforming the current (linear) economy in a more circular one, however, the way the biomass is produced, processed and BB products are produced are fundamental aspects to be properly assessed and monitored. This can be done using specific methodologies like LCA. Within this context the proposed MCI has to be seen as a complementary (quantitative) tool for further qualifying the sustainability of BB products and not as a substitute tool.

Declaration of interest

The author declares that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Metrics for quantifying the circularity of bioplastics: the case of bio-based and biodegradable mulch films

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Abstract

The concept of circularity and its quantification through the Material Circularity Indicator (MCI) is well established for traditional plastic products. In this paper a methodological approach for calculating the circularity of bio-based and biodegradable (BB) products is proposed and applied to BB mulch films. BB products are different from traditional products in as much as they are sourced and regenerated (recycled) not through technical cycles but the biological loop. The suggested method is an adaptation of the MCI where two major changes were made: (i) the mass of the bio-based component corresponds to the recycled material in input and (ii) the mass of the bio-based component leaving the system through composting or biodegradation in soil is accounted as recycled. The modified MCI supports the Eco-design of innovative BB products and allows for the comparison of their circularity taking into account the biological source and the expected end of life process such as biodegradation. To demonstrate the adaptation, the method has been applied to BB mulch film. Results showed that the MCI of a biodegradable mulch film, characterized by an average bio-based feedstock content of 30% is 0.37 ± 0.04 in a 0-1 scale. For BB mulch film, the amount of bio-based feedstock is the most sensitive factor and controls linearly the value of the MCI.

Keywords: circularity indicators, circular economy, bioplastics, biodegradable mulch film, bio-based product, biodegradation

Abbreviations	
BB	Biodegradable and bio-based
CE	Circular Economy
d.m.	Dry matter
EMF	Ellen MacArthur Foundation
LCA	Life Cycle Assessment
LDPE	Low-Density Poly-Ethylene
MCI	Material Circularity Indicator
NRF	Non-Restorative Flows
PBAT	Polybutylene adipate terephthalate
PE	Poly-Ethylene
PLA	Polylactic acid
PHB	Poly hydroxy butyrate

1 Introduction

To overcome today's unsustainable model of 'take-make-dispose' and its related risks such as hikes in raw material prices, pressures on the environment, shortage of global resources and waste sinks, a circular approach needs to be applied. It is a new regenerative economic view, based on a balance between economy, environment and society, a total resource efficiency and a Zero Emission Strategy that aims to maximize products value with zero, or minimal, environmental impact (Ghisellini et al., 2016) . Together with structural changes in environmental legislation, new logistics, technologies and sharing schemes, the Circular Economy (CE) approach which is regenerative by design, aims at closing materials loops, *i.e.* at reducing virgin materials input and waste output.

In December 2015, the European Commission developed an Action Plan for Circular Economy (European Commission, 2015), where plastic was considered a priority to be tackled. In January 2018, an *EU Plastic Strategy* (European Commission, 2018) was adopted, in order to react to the increasing environmental problems concerning plastic production, consumption, use and disposal along the same lines of the CE approach. Two fundamental steps to increase the circularity of different plastic products are (i) the abandonment of fossil fuels, *i.e.* currently 90% of the plastic is produced by virgin petroleum-based feedstock (Ellen MacArthur Foundation, 2017), and (ii) the development of easily recyclable products which are recycled. Today, in EU the share of plastics collected for recycling is 30% while the use of recycled plastics is just 6% (European Commission, 2018).

Biodegradable and bio-based (BB) plastics are spreading across markets (Institute for Bioplastics and Biocomposites, 2018) as a valid contribution to meet CE aims and principles. This is true as long as the supply of renewable raw materials, generally from agriculture, is based on a sustainable approach and the conversion processes along the

supply chain are efficient and highly integrated in a Life Cycle Assessment (LCA) perspective (EPLCA – European Platform on LCA). While traditional plastics can be mechanically recycled or incinerated with energy recovery, BB plastic products offer new recycling routes in waste management, due to their biodegradability. Organic recycling (through composting or anaerobic digestion) or in the case of specific applications such as agricultural mulch films, biodegradation in the environment, offer additional recovery options resulting in less wastes and less contamination of soil by plastic residues (Razza et al., 2012; Lange, B., 2016). An extensive literature review about the potentialities and benefits of renewable and compostable bioplastics, encompassing market perspective, applications, economic effects etc. can be found here: (BBIA; European Bioplastics).

Nevertheless, the research and development of innovative products, such as the BB products, implies the development of methodologies and metrics capable of measuring their circularity. Without this it is not possible to achieve measurable results and improving actions, as well as provide unequivocal references for comparisons of products of the same type/category. In 2015 the Material Circularity Indicator (MCI) was developed (Ellen MacArthur Foundation & Granta Design, 2015) which aims to quantify the regeneration of a product's material flow and is considered one of the few, among sixteen CE indexes suiting a micro-scale assessment of circularity at product or company level (Lonca et al., 2018). However, it focuses solely on technical cycles and recycled materials. Furthermore, recovery and recycling through the biological cycle offered by industrial composting, anaerobic digestion or biodegradation in natural environments are not considered as end of life options. In order to apply the MCI system to BB plastic products, the development of an enhanced methodology is necessary.

The approach proposed by the authors allows to quantify the circularity of BB plastic products and to make comparisons with equivalent traditional plastic products. To

demonstrate the applicability of the proposed method a computational example for mulch film products is provided. In so doing so, the paper aims at contributing to the Eco-design of these innovative products.

1.1 The case study of mulch films

Plastic mulch films represent an important agronomical technique well established for the production of many crops thanks to numerous agronomical advantages such as: increased yield and higher quality of productions (Steinmetz et al., 2016) ; weed control and reduced use of pesticides; early crop production and reduced soil moisture loss (Briassoulis and Giannoulis, 2018). As a consequence, the plastic films consumption has increased year-by-year, reaching a current global market estimated at 1.4 millions of tonnes , mainly in Asia (Briassoulis and Giannoulis, 2018; Mormile et al., 2017) , and covering 80,000 km^2 of agricultural surface (0.6% of the global arable land). The mulch film market in Europe is estimated by Agriculture Plastic & Environment and by the European Bioplastic Associations at 76-80 kt. The most used raw material is Poly-Ethylene (PE) in its different forms, due to its processability, chemical resistance, high durability and flexibility (Kasirajan and Ngouajio, 2012; Plasticulture, 2016 and 2018; Shen, M. et al., 2019; Wen, X. et al., 2018).

Despite these benefits, manifold environmental and agronomic problems have been pointed out. After its useful life – which in general does not exceed 1 to 3 months – the mulch film has to be removed and properly disposed of, a time-consuming (about 16 hours per hectare) and costly procedure (Scaringelli, M., 2016; Briassoulis, D., 2013). The recovered film is usually heavily contaminated with soil and organic residues, making mechanical recycling technically difficult and not a cost-efficient solution (Briassoulis et al., 2018; Figuier, 2016; De Lèpinau, Arbenz, 2016). The most common end of life of collected films in Europe is still landfilling (about 50%), followed by energy recovering

and finally mechanical recycling (Le Moine, 2014). Recent Chinese prohibition (January 2018) to import different types of wastes is heavily impacting the European agricultural plastic waste management, highlighting the difficulty in properly recycling this type of plastics (Tamma, 2018). Plastic films may not be properly collected and recycled but disposed of by burning in the field or by uncontrolled landfilling or left directly in the (agricultural) soils, causing serious environmental concerns. An example is the "White pollution" phenomena described in the Xinjiang Autonomous Region (China), in which the residual plastic film can reach 200 kg/ha in the top soil with detrimental effects on soils' quality, health and fertility (Liu, He, & Yan, 2014; Gao *et al.*, 2019; Steinmetz *et al.*, 2016).

As a reaction, there has been significant research into novel materials especially related to biodegradable and bio-based (BB) mulch films, which enable an effective biodegradation in soil and provide comparable agronomical performances (Touchaleaume et al., 2016). The term "bio-mulch film" brings together several types of both bio-based and fossil oil-based biodegradable polymers and blends of them, such as polylactic acid (PLA), polybutylene adipate co-terephthalate (PBAT), starch-based polymer blends or copolymers. They biodegrade when exposed to bioactive environments such as soil and compost (Kasirajan et al., 2012) which means that they can be left *in situ* to be fully biodegraded after being used. Clearly the biodegradation rate of biodegradable bioplastics is influenced by the environmental conditions such as the types of available bacteria, fungi thus specific enzymes namely native microflora (Pico, Y. et al., 2019). However their intrinsic biodegradability allow the complete biodegradation with times similar to natural polymers such as cellulose used as reference by the relevant standards and certification schemes.

The EN 17033:2018 is a new European Norm (standard) concerning “Plastics - Biodegradable mulch films for use in agriculture and horticulture - Requirements and test methods”, which sets the necessary tests and limits to define biodegradability, performances and environmental impacts of BB much films. The material is considered completely biodegradable if it achieves a complete biodegradation (absolute or relative to the reference material) in a test period no longer than 24 months (mineralization into CO_2). Additionally, a control of constituents (such as metals) and eco-toxicity testing (acute and chronic toxicity tests on plant growth, earthworm; nitrification inhibition test with soil microorganisms) were required. A certified mulch film guarantees that the product will completely biodegrade in the soil without adversely impacting on the environment.

1.2 Goal of the paper

The goal of the paper is to provide a general and common metric to measure the circularity of a bio-based and biodegradable (BB) product and to apply the methodology at product level to a category of products, namely bio-based and biodegradable mulch films.

2 Materials and Methods

2.1 MCI accounting according to the EMF methodology

The Material Circularity Indicator (MCI), according to the Ellen MacArthur Foundation (EMF) methodology (Ellen MacArthur Foundation & Granta Design, 2015), is a number that can range from 0 (pure linearity) to 1 (pure circularity). A purely linear production provides for the exclusive use of virgin raw materials that turn into waste at the end of the use phase of the product. Vice-versa, pure circularity includes the use of recycled materials and does not produce wastes (regenerative streams). Circularity can be achieved in different ways: as for the purpose of this paper, only recycling will be considered since

reuse is not an option for thin biodegradable mulch films. Since the method considers only mass flows, the recycling corresponds to the recovery of materials for the original purpose or for other purposes and excludes energy recovery, considered as a loss of materials equal to landfill disposal. The materials recovered feed back into the process as recycled feedstock.

The MCI methodology differentiates 'technical cycles' from 'biological cycles', modelling only the former. The first contains products and materials re-entering into the system (market) with the highest possible qualities and for as long as possible (thanks to reuse, repair, refurbishment and recycling) and the latter includes biological materials used in cascade until their restoration into the biosphere and the re-constitution of natural resources.

The material flows associated to the production of a generic technical cycle from non-renewable sources are summarized in Figure 1. The dashed lines indicate that recycled feedstock does not have to be sourced from the same product but can be acquired on the market. With reference to Figure 1, the list of the parameters used in the EMF methodology is reported in Table 1, while the equations relevant for the analysis carried out in this paper are described in the following sections (Table 2, Chapter 2.2).

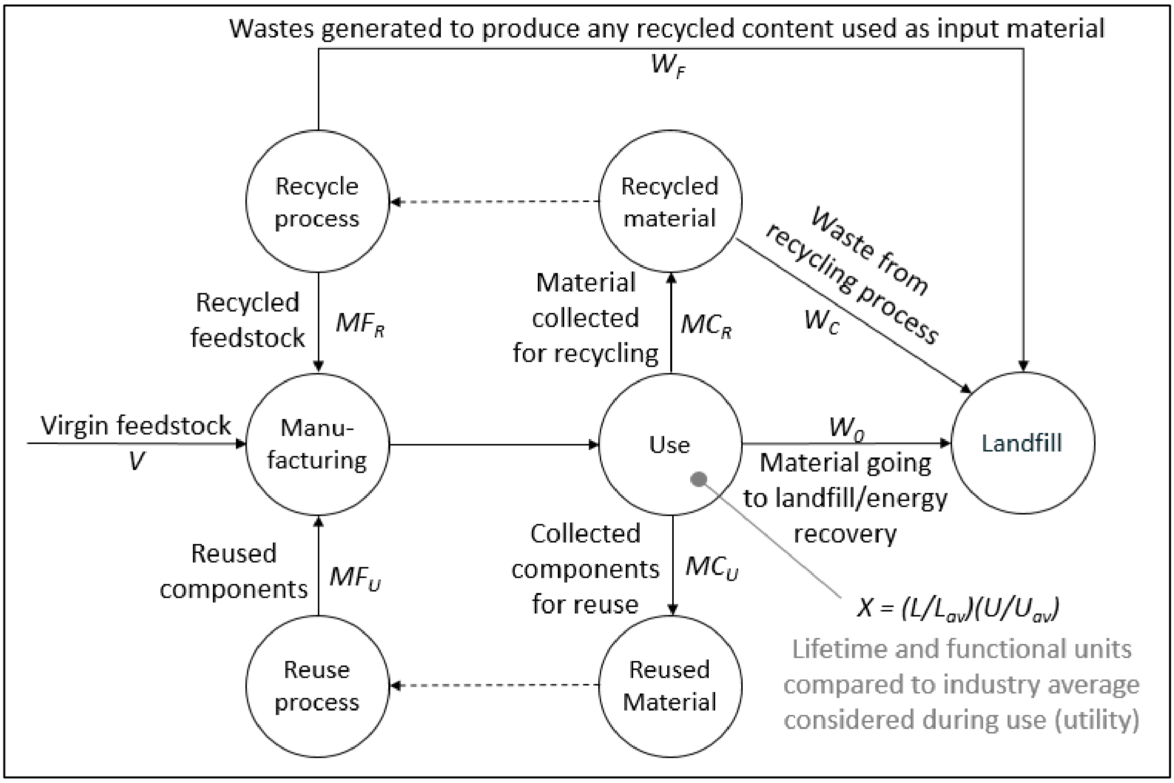

Figure 1: *Diagram of material flows and associated variables of a generic product (modified from Ellen MacArthur Foundation & Granta Design, 2015).*

Table 1: *Parameters and relative definitions used in the EMF methodology.*

Parameter	Definition
M	Total mass of the product
F_R	Fraction of mass of a product's feedstock from recycled sources
F_U	Fraction of mass of a product's feedstock from reused sources
V	Mass of virgin feedstock used in a product
C_R	Fraction of mass of a product being collected to go into a recycling process
C_U	Fraction of mass of a product going into component reuse
E_C	Efficiency of the recycling process used for the portion collected for recycling
E_F	Efficiency of the recycling process used to produce recycled feedstock for a product

$\boldsymbol{W}$	Total mass of unrecoverable waste associated with a product
$\boldsymbol{W_0}$	Mass of unrecoverable waste (landfill, waste to energy and any other type of process where the materials are no longer recoverable)
$\boldsymbol{W_C}$	Mass of unrecoverable waste generated in the process of recycling parts of a product (after use)
$\boldsymbol{W_F}$	Mass of unrecoverable waste generated when producing recycled feedstock for a product
$\boldsymbol{X}$	Utility of a product, calculated as $X = (L/L_{av})(U/U_{av})$
$\boldsymbol{L}$	Actual average lifetime of a product
$\boldsymbol{L_{av}}$	Actual average lifetime of an industry-average product of the same type
$\boldsymbol{U}$	Actual average number of functional units achieved during the use phase of a product
$\boldsymbol{U_{av}}$	Actual average number of functional units achieved during the use phase of an industry-average product of the same type

The Material Circularity Indicator is determined as follows: , where *LFI* is the Linear Flow Index measuring the flows of virgin materials and unrecoverable wastes associated to the examined product.

A function of the utility, – , is used to correct the *LFI*. The function *F* is chosen in such a way that improvements of the utility of a product (e.g., by using it longer) have the same impact on its MCI as a reuse of components, leading to the same amount of reduction of virgin material use and unrecoverable waste. Setting a = 0.9, MCI takes, by convention, the value 0.1 for a fully linear product (*i.e.*, $LFI = 1$) whose utility equals the industry average (*i.e.*, $X = 1$). This leaves some margin to distinguish between processes with a high linearity but different utilities.

2.2 MCI accounting for bio-based and biodegradable (BB) products

To apply the EMF methodology to BB products, formulas and flows (Figure 1 and Figure 2) are adapted as it follows:

1. The fraction of the recycled feedstock, F_R, corresponds to the share of the bio-based feedstock content in the final BB product, $F_{R(i)}$. It is the ratio of the d.m. amount of bio-based feedstock per d.m. amount of the total mass of BB product (EN 16785-2:2016).
2. The fraction of restorative mass going into a recycling process, C_R, corresponds to the share of bio-based feedstock content in the BB product biologically recovered (*e.g.* through composting) or biodegraded in the natural environment, as it happens for specific applications (e.g. biodegradable mulch film, *etc.*). It is the ratio of the d.m. amount of bio-based feedstock per d.m. amount of the total mass of BB product that is biologically recycled.

The modified scheme is shown in Figure 2. Table 2 lists the formulas as adapted to BB products.

Table 2: *List of formulas as developed by EMF methodology compared to the proposed adaptation to BB products.*

EMF methodology	Adaptation to BB products

The mass of fossil-based feedstock which may be contained in BB products (V) is obtained as a difference of the total mass (M) minus the bio-based fraction; in this case the F_R in the EMF methodology corresponds to the sum of the fractions of all the bio-based feedstock/s used in manufacturing the BB product. Therefore, is the total bio-based feedstock mass in the product. In single-use products, such as mulch films, reuse is not considered for BB products, so that $F_U = C_U = 0$.

W_F is the total amount of unrecoverable waste associated to the production of bio-based feedstock used to produce BB products (*i.e.* the amount of uncoverable waste per unit of BB product). Bio-based feedstocks such as starch, PLA, PHB etc. generate non-restorative flows which can be quantified. Such unrecoverable waste correspond to $R_{(i)}$, the specific amount of waste generated within cradle-to-gate boundaries per unit of bio-based feedstock going into manufacturing, and it is estimated through LCA studies. Thus all inputs from growth and harvesting phases and the related wastes generated by fertilisers and pesticides are here accounted. $R_{(i)}$ can be easily found in specific literature or life cycle inventories (LCI) present in LCA databases. In the calculation of W_F, also the efficiency of manufacturing process of BB products E_P is considered, as the ratio of the

overall bio-based feedstock content in the final BB product to the bio-based feedstock in input to the manufacturing process.

The material flows associated to the production of a generic BB product are summarized in Figure 2.

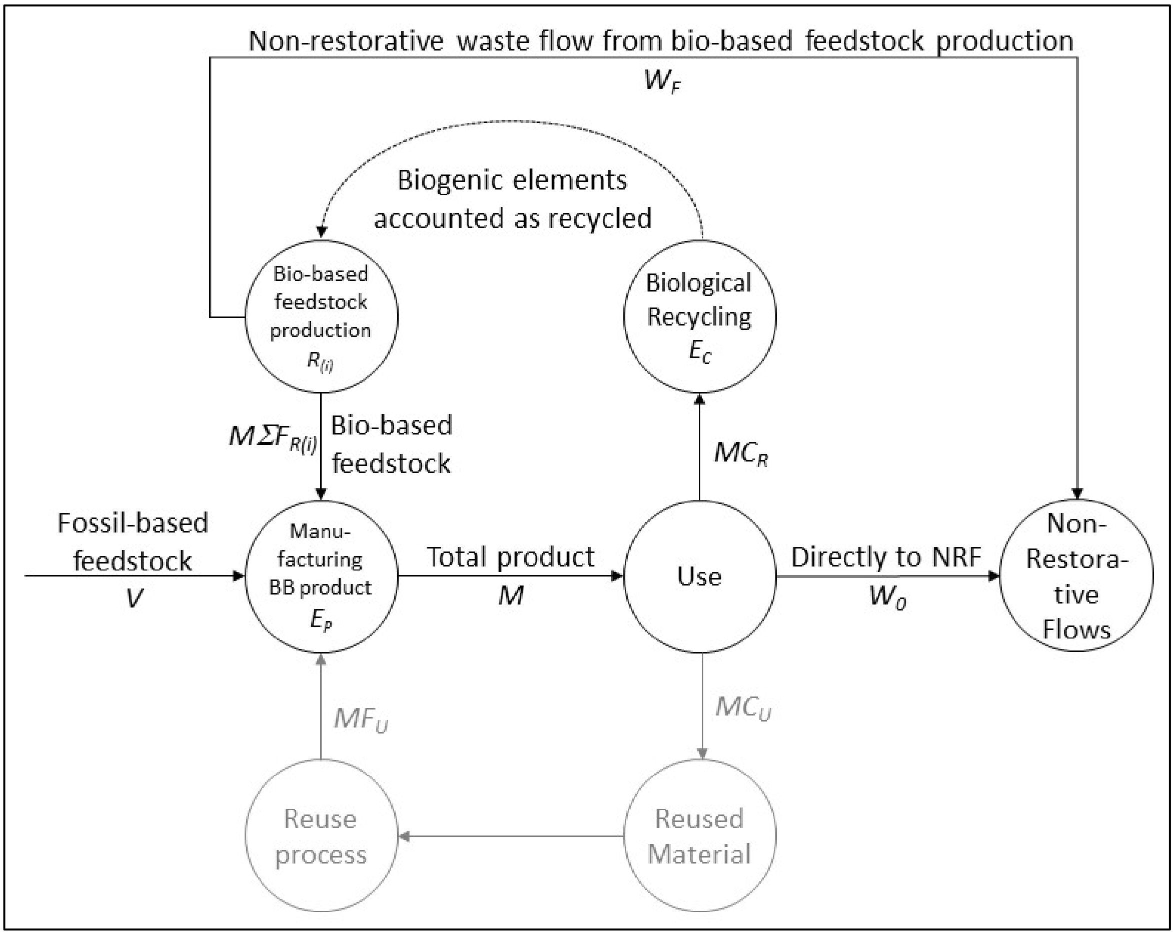

Figure 2: *Description of material flows adaptation to BB products; in this paper, the reuse flow is out of scope* ($C_U = F_U = 0$).

The biodegradation of bio-based feedstock does not imply the generation of waste W_C as it occurs in a standard mechanical recycling process. This implies that C_R and E_C (*i.e.* the efficiency of the biodegradation process) equal to 1. Indeed, a BB raw material, sent to biological treatment (composting) or biodegraded in a natural environment, is fully transformed in its chemical elements (C, H and O mainly) derived from the decomposition of complex molecules (polymers) without the release of waste (Witt et al., 2001; Marten et

al., 2003; Eubeler et al., 2010; BASF, 2018; Institute of Bioplastics and Biocomposites, 2018; OWS, 2018; Zumstein et al., 2018). These natural elements return into the environment and are then available in the respective biogeochemical cycles. The (biodegradable) fossil portion behaves as well; consequently, $W_C = 0$.

Nevertheless, the fossil-based feedstock cannot be considered as a regenerative circular feedstock, since it derives from carbon stored for millions of years and extracted by man, not being part of the active and fast biogeochemical carbon cycle. This is accounted in the quantification of W_0, the mass of unrecoverable waste from use (*i.e.* the linear stream going to landfill or incineration, the Non-Restorative Flows, NRF), as , the total amount of fossil-based feedstock.

Since W_F and W_C are associated to complete different processes and W_C is always equal zero, the double counting issue does not occur and the quantification of *W* and *LFI* is modified as reported in Table 2.

2.3 MCI calculation for mulch films: scope, inventory and assumptions

The new formulas reported in Table 2 were applied to a single use product namely a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1.

In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the BB film has been arbitrarily assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e. 23% of starch, $F_{(S)}$, and 7% of a bio-based additive, $F_{(BA)}$), while the rest was assumed to consist of fossil feedstock (

Figure *3*). Since a generalized approach was used and no primary data were implemented, the information were extrapolated from literature (Institute of Bioplastics and Biocomposites, 2018); the main characteristics of the two examined products are presented in Table 3.

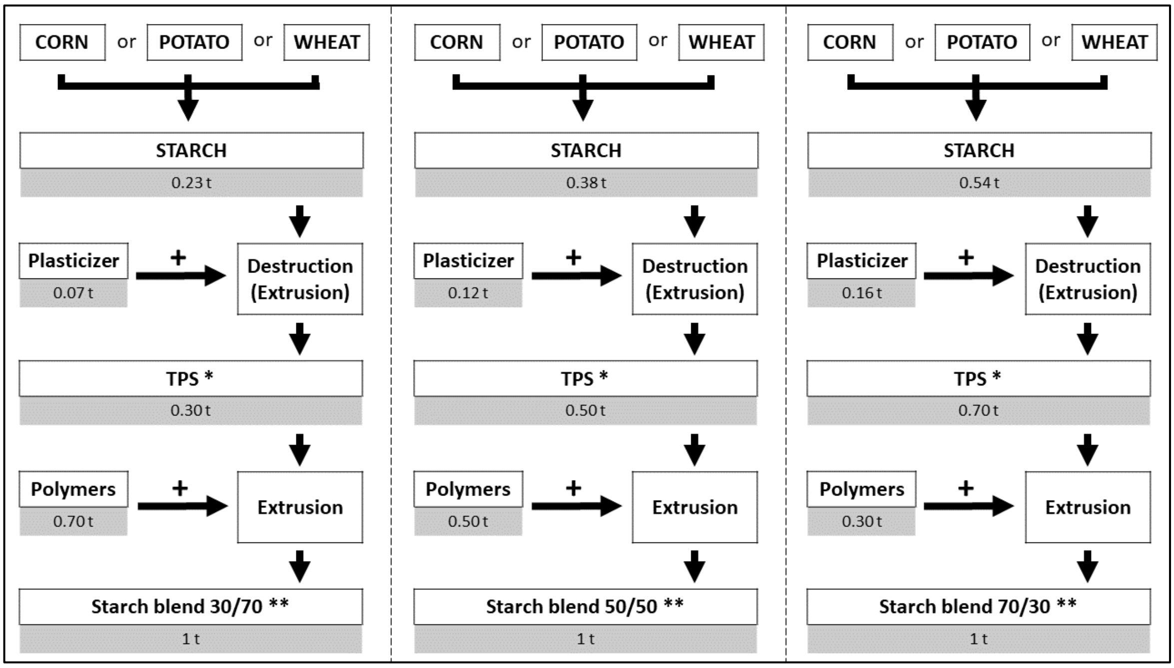

Figure 3: *Examples of hypothetical bio-based polymers; in this paper, the first option on the left (starch blend 30/70) has been chosen for carrying out the numerical MCI calculation (working hypothesis). The figure considers a 100%-efficiency in every phase of production, so that the residues are equal to zero; the same assumption is done in this paper. *TPS (Thermoplastic starch), starch content 75%; **Ratio TPS/Polymer; modified from Institute of Bioplastics and Biocomposites, 2018.*

Table 3: Key features representative of the BB mulch films.

	BB mulch film
Material	30% bio-based feedstock (23% starch + 7% bio-based additive) + 70% fossil feedstock
Thickness (µm)	12
Density (g/cm³)	1.25
Weight (g/m²)	15.2
Functional unit (the covering of the agricultural land)	6000 m²/ha (the actual mulched soil in a hectare is generally equal to the 60% of the total area; Malinconico, 2017)

In the calculation of MCI for the BB mulch film, the adapted formulas were used together with assumptions. As stated before, BB mulch films are blends of bio-based and fossil based feedstocks (in the specific case, 30% and 70% respectively). Unlike the LDPE mulch film that has to be removed and disposed of, the BB mulch film is left in soil where it undergoes an ultimate biodegradation (so that $C_R = 1$) with no waste (so that $E_C = 1$), in respect of the specific standard EN 17033:2018. As a result of polymers' decomposition, the derived (biogenic) C, H and O finally return into biosphere (atmosphere, microorganism biomass, organic material pool) (OWS, 2018), and back into biogeochemical cycles in a relatively short time ("Biogenic elements accounted as recycled" in Figure 2), with the exception of humified compounds. Actually, also C, H and O deriving from fossil-based sources undergo biodegradation (Zumstein, M.T., 2018) but they are not considered as a regenerative flow ("Waste from non-restorative flow" in *Figure 2*) and their "wastes" are indeed calculated in W_0.

Applying a conservative approach, W_F, the waste generated by the production of each bio-based feedstock, is quantified considering a "cradle to gate" LCA study. The estimated solid wastes $R_{(i)}$ for the presented case study are related to the production of starch ($F_{(S)}$),

with an amount $R_{(S)}$ of 0.014 kg of waste per kg of renewable feedstock (source: personal communication A. Novelli), and to the production of the bio-based additive ($F_{(BA)}$), with $R_{(BA)}$ equals to 0.025 kg waste/kg renewable feedstock (US-LCI database). As assumed in

Figure *3*, the production efficiency of BB product E_P (how much bio-based feedstock is needed for every unit of BB product) is estimated equal to 1 and no unrecoverable wastes are generated by the process.

In addition, an explorative sensitivity analysis has been performed regarding exclusively the amount of bio-based feedstock content of the BB mulch film, *i.e.* (*i.e.*, $F_{(S)}$ + $F_{(BA)}$), as shown in Figure 4 (Chapter 3). Considering the characteristics of the films (weight, g/m^2, or thickness, μm, and density, g/cm^3) and the relative functional unit (6000 m^2/ha, Table 3), it is possible to calculate a mass, *M*, that is 90 kg/ha for the BB one. Once calculated the masses, the formulas reported in Table 2 (Chapter 2.2) are applied. Results are shown in Table 4.

2.4 Sensitivity analysis

A sensitivity analysis was conducted for BB mulch film to examine the effects of changing the main variables. Given a non-linear dependence of results on parameter values, a Monte Carlo approach (see, *e.g.*, Lloyd and Ries, 2008) has been adopted. The model has been implemented using specifically written routines in the C++ programming language. The model was run with 100,000 events for BB mulch film, where the value of each parameter has been randomly chosen following a Gaussian distribution with a standard deviation within a range of possible and realistic values (Table 5 and **Error! Reference source not found.**; Figure 5 and Figure 6).

3 Results

Figure 4 shows how the value of the MCI varies according to the percentage variation of the bio-based feedstock in the total mass of the product.

Table 4: *Resulting parameters in the calculation of MCI for BB mulch film.*

Parameter	**BB mulch film**

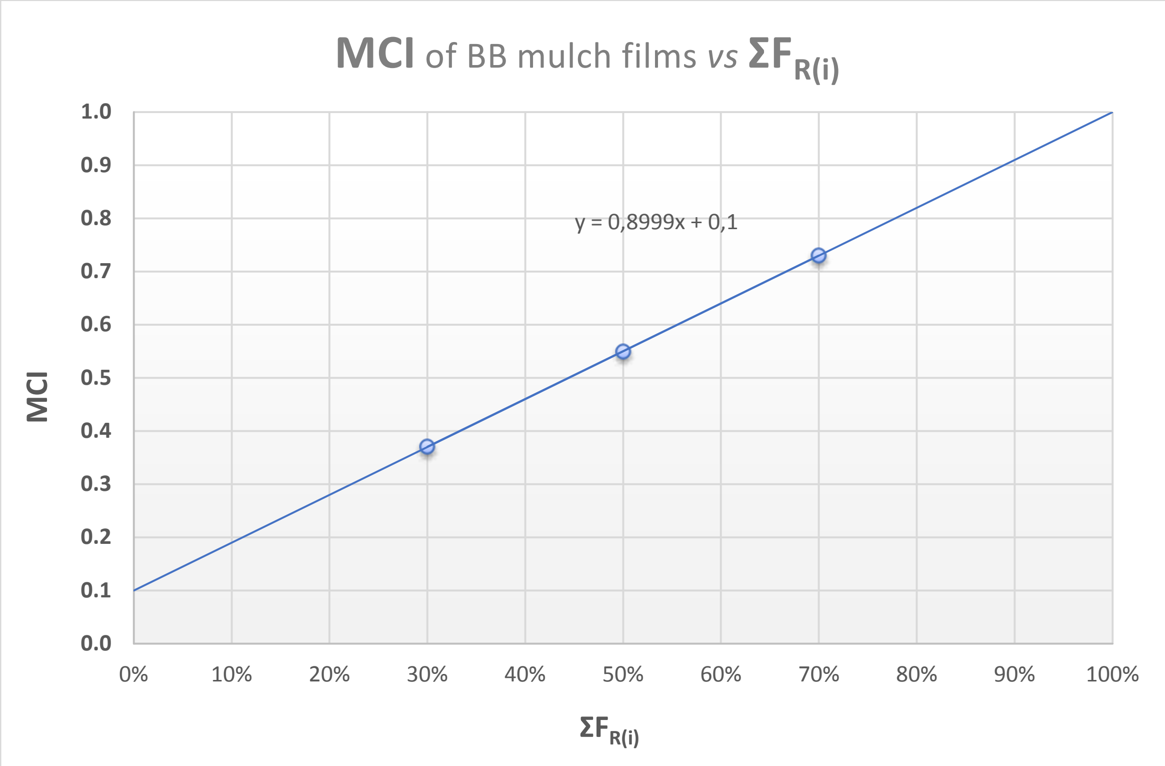

Figure 4: *MCI as a function of the amount of bio-based feedstock/s in the BB mulch film* $\Sigma F_{R(i)}$*, expressed as the percentage of all the bio-based feedstock/s of the mulch film on dry mass basis (X-axis). The dots correspond to the three different hypothetical bioplastic compositions of Figure 3.*

3.1 Sensitivity analysis

The results of the sensitivity analysis are presented in the followings Table 5 and Figure 5 and Figure 6. The accuracy band is a fraction of the average and corresponds to a probability of 95%. It has been chosen in order to be representative of the variability of the product category, the BB mulch films. The simulation can thus be regarded as a system composed by a high number of companies, each producing films with different characteristics, that are accounted for in the accuracy band.

Table 5: *Parameters used for the sensitivity analysis of the BB mulch film. (**) The Accuracy Band is defined as twice the standard deviation of the distribution.*

Variable name	Average	Accuracy Band (**)	Unit
M	1000.00	0%	kg
$\mathbf{F_{(S)}/F_{(BA)}}$	3.29	10%	fraction
$\mathbf{F_{(S)} + F_{(BA)}}$	0.30	30%	fraction
$\mathbf{F_{U}}$	0.00	0%	fraction
$\mathbf{C_{U}}$	0.00	0%	fraction
$\mathbf{R_{(S)}}$	0.014	100%	fraction
$\mathbf{R_{(BA)}}$	0.025	100%	fraction
$\mathbf{E_{C}}$	1	0%	fraction
$\mathbf{E_{P}}$	0.95	10%	fraction
$\mathbf{C_{R}}$	1.00	0%	fraction

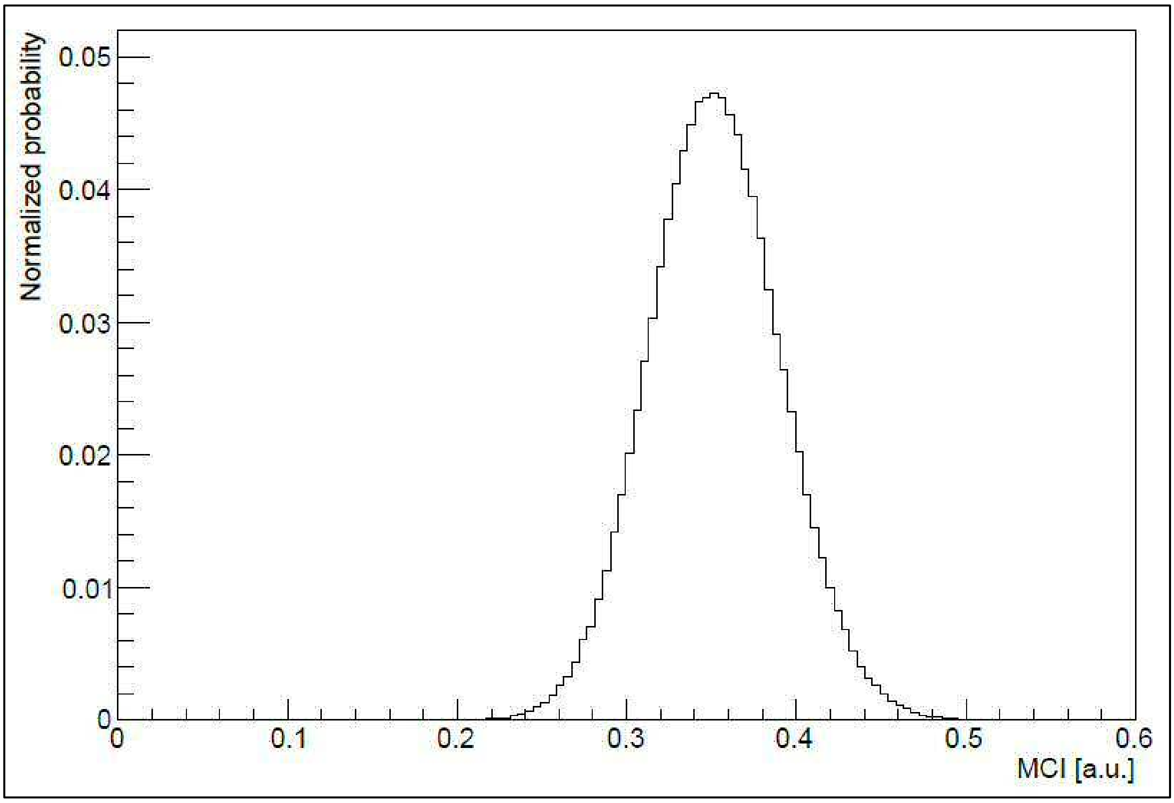

Figure 5: *Resulting distribution of MCI values for BB mulch film.*

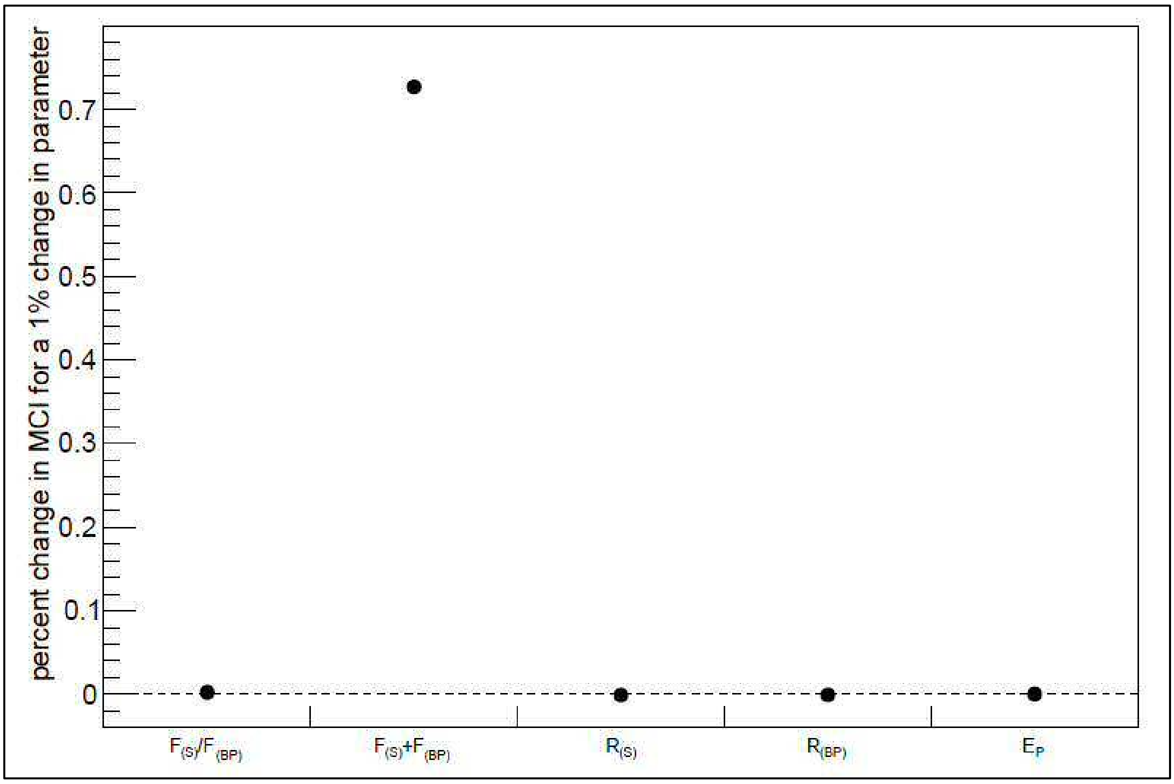

Figure 6: *The most sensitive and relevant parameters in the calculation of the MCI of the BB mulch films.*

4 Discussion

This work applies the principles of the EMF methodology into BB products so as to define common metrics for calculating their circularity. By doing so it proposes some substantial changes to the EMF methodology but still coherent with the overall methodological framework. Such changes should be seen as a generalisation of the methodology provided the following rules are applied:

(1) fossil-based feedstocks or component materials embodied in the BB products whatever is the final disposal (even biological recycling) shall be considered as non-restorative;

(2) bio-based component materials embodied in the BB product that go to biological recycling like composting, or biodegrade in the environment (i.e. BB mulch film) shall be considered restorative as long as they flow through the biosphere safely, without any harm to the environment (e.g. no toxicity effects).

(3) bio-based component materials embodied in the BB product that go to incineration and landfill shall be considered as non-restorative;

The justification of these rules is described in the following.

Fossil-based component materials in the product derive from deposits where they remained stocked for a geological time scale. Once the product is mineralised, its fossil-based portion will be accounted as non-regenerative and therefore linear, due to its origin (Joos et al., 2013). This is true, even if fossil carbon, for example, will re-enter biological cycles, like CO_2 in the atmosphere and other streams, since both fossil-based and bio-based component materials will physically and chemically behave the same, once biodegraded. However, the source of the bio-based carbon was circular before its use (concept of "carbon neutrality", equilibrium between the biogenic carbon released and the carbon absorbed by plants) and will maintain its circularity provided that the carbon is released into the atmosphere at the same rate. The reason has its origin in the EMF general provisions stating that “biologically sourced materials can only be considered part of a Circular Economy if materials are not used faster than they can be restored naturally” (Ellen MacArthur Foundation & Granta Design, 2015). If BB products are incinerated, the bio-based components are still considered linear, maintaining consistency with EMF principles. Basically, a complete circularity for a BB product is satisfied when its renewable components are 100% bio-based and they go 100% to biological recycling or biodegraded in the environment (for specific application like mulch film).

As for provision (3), a material health rule has its origin in manyfold normative definitions of the CE. In addition, the EMF definition of biological cycles is that of non-toxic materials which are restored into the biosphere and the CE is defined as such if it can “eliminate the use of toxic chemicals”. The need of a safety clause has been reviewed under many aspects by Verberne (2016) and can be put as a postulate of the restoration principle: if a flow is toxic it cannot be defined restorative. This is also at the core of the REACH Regulation (EC 1907/2006). In the specific case, the material complies with the standard EN 17033-2018 certifying that no harm is caused to a) all relevant organism groups as plants, invertebrates (e.g. earthworm) and microorganisms, b) important ecological processes maintaining soil functions, c) all relevant exposure pathways as soil pore water, soil pore air and soil material.

A comprehensive approach for MCI calculation should also include non-restorative flows generated at upstream level like biomass growth, in the specific case corn, and biomass conversion processes like starch extraction and refining. Specifically these non-restorative flows correspond to the overall non-recyclable wastes associated to the bio-based feedstock supply thus non-recyclable waste from fertilizer and pesticide production, non-recyclable scraps from conversion processes, etc. In this study such flows of non-restorative waste coming from upstream manufacturing operations were included for the

bio-based feedstocks ($R_{(i)}$) used in manufacturing the BB mulch film applying "cradle to gate" LCA methodology. However, we observed that the inclusion of upstream unrecoverable waste does not significantly influence the MCI results in the chosen case study, since the respective amounts are small. The specific unrecoverable waste for starch and bio-based additive (i.e. kg of waste/kg of bio-based feedstock) were estimated at 0.014 and 0.025, respectively.

The resulting MCI for the 30/70 blend of the BB mulch film is equal to 0.37 in a 0-1 scale and its circularity is linearly linked to the amount of bio-based feedstock used according to the equation $y = 0.89x + 0.1$, where y is the MCI and x is the bio-based feedstock content, therefore the amount of recycled feedstock or (renewable) bio-based feedstock in input is decisive.

Apart from the specific application analysed in this paper, the proposed MCI method can be easily applied and calculated for any kind of BB product as long as the following information are available:

- The bio-based feedstock content, determined according to the standard EN 16785-2:2016, if the composition is known, or directly provided by the BB product manufacturer.
- The End of Life scenario of the studied BB product (real or hypothetical).
- The amount of un-recoverable waste associated to the production of bio-based feedstock contained in the BB product. They can be derived from LCA databases or other specific sources.

5 Conclusions

Bioplastic market is steadily increasing. The value proposition of bio-based and biodegradable products is linked to:

1. the use of renewable feedstock (like starch and its derivates) instead of fossil oil or natural gas;
2. the waste recovery through biological recycling, thanks to their ability to biodegrade in composting facilities or in soil (*e.g.* biodegradable mulch film).

The Material Circularity Indicator (MCI), developed by the EMF, is a metric for quantifying "how much" a product is circular (MCI = 0, fully-linear product; MCI = 1, completely circular product) thus it represents a valuable tool for product eco-design purposes. However, it focuses solely on technical materials, mechanically recycled or reused, leaving out bio-based feedstocks and related biological treatments such as composting. Without common metrics it is not possible to pursue concrete actions, to achieve measurable results and to provide unequivocal references for all products. This research work aims at filling this gap through the development of a methodology coherent with EMF MCI methodology but able to catch the specificities of bio-based and biodegradable products and provide metrics for those innovative products. Direct uses are: (i) supporting the eco-design of innovative bio-based products and (ii) comparing the MCI of BB products with MCI of traditional products (e.g. fossil based).

The proposed method has been applied to a real case study (*i.e.* biodegradable mulch film) providing quantitative metrics about its circularity. Specifically considering a bio-based feedstock content of 30%, the correspondent MCI is 0.37 in a 0-1 scale and its circularity is heavily linked to the bio-based feedstock content according to this relation: $MCI_{(BB\ mulch\ film)} = 0.89 * bio\text{-}based\ feedstock + 0.1$.

The MCI is a key performance indicator to develop more circular products, in line with the Circular Economy principles like the use of renewable materials and the reduction of the amount of not recoverable waste. MCI will support the development of innovative products just based on these two important characteristics specific for each BB

product/application and end of life scenario Bioeconomy, thus also BB products, can provide valuable insights in transforming the current (linear) economy in a more circular one, however, the way the biomass is produced, processed and BB products are produced are fundamental aspects to be properly assessed and monitored. This can be done using specific methodologies like LCA. Within this context the proposed MCI has to be seen as a complementary (quantitative) tool for further qualifying the sustainability of BB products and not as a substitute tool.

Declaration of interest

The author declares that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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HIGHLIGHTS

1. A modification of the MacArthur methodology on product circularity (i.e. Material Circularity Indicator MCI) has been developed to make it applicable to bio-based and biodegradable (BB) products.
2. The proposed metric has been applied to a specific case study: the bio-based and biodegradable mulch film.
3. Results show that a biodegradable mulch film with a 30% of bio-based feedstock content is characterized by a MCI of 0.37 ± 0.04 in a 0-1 scale.
4. For a BB mulch film the amount of bio-based feedstock is the most sensitive factor and controls linearly the value of the MCI.

REVISED HIGHLIGHTS

5. A MCI methodology suitable for Bio-based and Biodegradable (BB) products has been developed.
6. The proposed metric has been applied to a specific case study: BB mulch film.
7. BB mulch film with a 30% of renewable feedstock is characterized by a MCI of 0.37 ± 0.04 in a 0-1 scale.
8. The amount of renewable feedstock is the most sensitive factor of the MCI

***conflict of Interest Statement**

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Author Contributions Section

Francesco Razza: Conceptualization, Methodology, Writing - original draft, Writing - Review & Editing, Data Curation, Investigation, Validation, Supervision

Cristiana Briani: Writing - Original Draft, Validation

Tony Breton: Writing - Original Draft, Supervision

Diego Marazza: Writing - Original Draft, Data curation, Validation

Dear Reviewers,
Many thanks for your time. The table below provides our replies to your further comments and the description of the changes made on the paper for each raised point. Many thanks again to all of you for the valuable comments and suggestions that allow us to further improve the work.

n	Reviewers'comments	Revisions made in the paper
	Reviewer #1	
1	All the issues mentioned by the reviewers have been addressed, and the paper quality has been greatly improved. Now, the manuscript may be considered for acceptance.	Many thanks
	Reviewer #3	
2	The authors have prepared an extensive revision of the original manuscript and addressed the reviewers' comments in a satisfactory manner. I have one major and one minor comment at this stage, and recommend acceptance of the work. I do not need to see a possible further revision.	Many thanks
3	Major comment: I am not convinced that complex material interactions (fossil carbon biodegrading, harmful organic waste, bio-based material recycling, etc.) can be meaningfully represented in a single indicator such as the MCI or its derivatives. But that is something that the community should decide and not the reviewers, by taking up your work or not. But I ask you to add a short remark on the critique of the general usefulness of this indicator in the discussion section.	Actually, we fully agree with you. The MCI here proposed is meaningful for judging how much circular a bio-based and biodegradable product is only if the bio-based material/product does not cause toxic concerns or issues. This is our postulate reported in R396-406. That said we have further pointed out this very important aspect in the conclusion and made an addition in R468-471.
4	One minor comment remains: + L202 and other places: The abbreviation d.m. is not clear to me. Please spell out! Dry mass?	It stands for dry matter. On page 1 under "abbrevations" section is reported d.m. = Dry matter.
	Reviewer #4	
5	The authors have satisfactorily addressed the comments raised by the previous reviewers and appropriately modified the manuscript.	Many thanks
6	This work attempts to augment the MCI proposed by EMF. Although the need for the work is clear, however recently (in 2019) EMF has already proposed MCI for biological products. Hence, authors need to compare and contrast the MCI proposed in this work with EMF MCI for bio products.	Many thanks for this comment. Following your hint we have found that the EMF methodology has been recently changed https://www.ellenmacarthurfoundation.org/assets/downloads/ce100/MCI-SC-28Nov-2019-Master-MB-4.pdf however we would like to point out that our work started long before the changes of MCI and in an complete independently way. For the sake of clarity we here report the (documented) main stages of our original work followed by our proposal for handling this issue. Story of our paper. **2017**: preliminary idea of the methodology **2018**: the beta version of the methodology is presented within the third working group of the

		Italian *Circular Economy Stakeholder Platform* (ICESP www.icesp.it). On page 38 of the ICESP report (dated December 2018) here available https://www.icesp.it/landing/docs/gdl/gdl3/REPORT_GdL3%20Strumenti%20per%20la%20misurazione%20dell%E2%80%99economia%20circolare.pdf a brief description of the – not final- methodology is provided. Please note that the report is dated December 2018 and it was developed in the last four month period of 2018. **2019**: within StarBioPro project http://www.star-probio.eu/ thanks to the collaboration between Novamont and the University of Bologna (PhD D. Marazza and Prof. A. Contin) the methodology was further developed and improved till the present version. The first submission of the paper occurred the 31st of April 2019. At that time we were not aware about the EMF initiative about biological products so we wrote our paper blissfully unaware. That said, we have seen that some consideration of the recast EMF methodology are very close to what we proposed. As an example, - a principle "ensuring biological materials remain uncontaminated and biologically accessible" has been added - virgin material now considers the biological materials fraction in its formula - all formulas now include the contribution of biological materials - composting has been added as an end-of-life option. However, the recast MCI differs now from our proposal because it accounts for energy recovery of biological materials which can make the MCI of a BB product higher than what we propose. Other points are still open such as the demonstration that the feedstock has been extracted from "Sustained Production". To compare and defend our choices against the recast MCI would require to re-write almost completely sections 2 and 3, all figures, tables and formulas included. Section 4 ought to be extended and oriented to a comparison of our methodological proposal versus the recast MCI. We believe this makes the case for an additional, different paper, while the purpose of this paper is still justified. Indeed, we would like to remark that the new MCI does not provide any specific guidance on practical

		cases as we did for the biodegradable mulch film. For these reasons we believe our paper can give an important scientific contribution to the debate. We decided to add an addendum in the paper reciting as follows: While this paper was undergoing peer review the authors became aware that the EMF published an update of the MCI methodology (Ellen MacArthur Foundation & Granta Design, 2019) including the extension of it to include the treatment of biological materials. This update introduces new definitions and formulas. The authors believe that most of the changes regarding accounting are in the direction here proposed and that this study can contribute as an illustration on how the material circularity of a biological based material can be addressed in a real case study. Furthermore the authors would like to highlight that the proposed methodology started long before the EMF changes: specifically the original idea dated back to 2017 and a beta version of it - not as it is now - was presented in the middle of 2018 at the Italian Circular Economy Stakeholder Platform (ICESP www.icesp.it).
		Beyond the integrations described above we have further integrated the section "Acknowledgements" with the following text since, as described above, the final development and refinement of the methodology has been carried out within StarProBio project along with the project partner University of Bologna (PhD Diego Marazza). Added text The contents of the paper are part of the findings of the project STAR-ProBio. STAR-ProBio has received funding from the European Union's Horizon 2020 program research and innovation programme under grant agreement No. 727740

Metrics for quantifying the circularity of bioplastics: the case of bio-based and biodegradable mulch films

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Abstract

The concept of circularity and its quantification through the Material Circularity Indicator (MCI) is well established for traditional plastic products. In this paper a methodological approach for calculating the circularity of bio-based and biodegradable (BB) products is proposed and applied to BB mulch films. BB products are different from traditional products in as much as they are sourced and regenerated (recycled) not through technical cycles but the biological loop. The suggested method is an adaptation of the MCI where two major changes were made: (i) the mass of the bio-based component corresponds to the recycled material in input and (ii) the mass of the bio-based component leaving the system through composting or biodegradation in soil is accounted as recycled. The modified MCI supports the eEco-design of innovative BB products and allows for the comparison of their circularity taking into account the biological source and the expected end of life process such as biodegradation. To demonstrate the adaptation, the method has been applied to BB mulch films. Results showed that the MCI of a biodegradable mulch film, characterized by an average bio-based feedstock content of 30% is 0.37 ± 0.04 in a 0-1 scale. For BB mulch film, the amount of bio-based feedstock is the most sensitive factor and controls linearly the value of the MCI.

Keywords: circularity indicators, circular economy, bioplastics, biodegradable mulch film, bio-based product, biodegradation

Abbreviations	
BB	Biodegradable and bio-based
CE	Circular Economy
d.m.	Dry matter
EMF	Ellen MacArthur Foundation
LCA	Life Cycle Assessment
LDPE	Low-Density Poly-Ethylene
MCI	Material Circularity Indicator
NRF	Non-Restorative Flows
PBAT	Polybutylene adipate terephthalate
PE	Poly-Ethylene
PLA	Polylactic acid
PHB	Poly hydroxy butyrate

1 Introduction

To overcome today's unsustainable model of 'take-make-dispose' and its related risks such as hikes in raw material prices, pressures on the environment, shortage of global resources and waste sinks, a circular approach needs to be applied. It is a new regenerative economic view, based on a balance between economy, environment and society, a total resource efficiency and a Zero Emission Strategy that aims to maximize products value with zero, or minimal, environmental impact (Ghisellini et al., 2016) . Together with structural changes in environmental legislation, new logistics, technologies and sharing schemes, the Circular Economy (CE) approach which is regenerative by design, aims at closing materials loops, *i.e.* at reducing virgin materials input and waste output.

In December 2015, the European Commission developed an Action Plan for Circular Economy (European Commission, 2015), where plastic was considered a priority to be tackled. In January 2018, an *EU Plastic Strategy* (European Commission, 2018) was adopted, in order to react to the increasing environmental problems concerning plastic production, consumption, use and disposal along the same lines of the CE approach. Two fundamental steps to increase the circularity of different plastic products are (i) the abandonment of fossil fuels, *i.e.* currently 90% of the plastic is produced by virgin petroleum-based feedstock (Ellen MacArthur Foundation, 2017), and (ii) the development of easily recyclable products which are recycled. Today, in EU the share of plastics collected for recycling is 30% while the use of recycled plastics is just 6% (European Commission, 2018).

Biodegradable and bio-based (BB) plastics are spreading across markets (Institute for Bioplastics and Biocomposites, 2018) as a valid contribution to meet CE aims and principles. This is true as long as the supply of renewable raw materials, generally from agriculture, is based on a sustainable approach and the conversion processes along the

supply chain are efficient and highly integrated in a Life Cycle Assessment (LCA) perspective (EPLCA – European Platform on LCA). While traditional plastics can be mechanically recycled or incinerated with energy recovery, BB plastic products offer new recycling routes in waste management, due to their biodegradability. Organic recycling (through composting or anaerobic digestion) or in the case of specific applications such as agricultural mulch films, biodegradation in the environment, offer additional recovery options resulting in less wastes and less contamination of soil by plastic residues (Razza et al., 2012; Lange, B., 2016). An extensive literature review about the potentialities and benefits of renewable and compostable bioplastics, encompassing market perspective, applications, economic effects etc. can be found here: (BBIA; European Bioplastics).

Nevertheless, the research and development of innovative products, such as the BB products, implies the development of methodologies and metrics capable of measuring their circularity. Without this it is not possible to achieve measurable results and improving actions, as well as provide unequivocal references for comparisons of products of the same type/category. In 2015 the Material Circularity Indicator (MCI) was developed (Ellen MacArthur Foundation & Granta Design, 2015) which aims to quantify the regeneration of a product's material flow and is considered one of the few, among sixteen CE indexes suiting a micro-scale assessment of circularity at product or company level (Lonca et al., 2018). However, it focuses solely on technical cycles and recycled materials. Furthermore, recovery and recycling through the biological cycle offered by industrial composting, anaerobic digestion or biodegradation in natural environments are not considered as end of life options. In order to apply the MCI system to BB plastic products, the development of an enhanced methodology is necessary.

The approach proposed by the authors allows to quantify the circularity of BB plastic products and to make comparisons with equivalent traditional plastic products. To

demonstrate the applicability of the proposed method a computational example for mulch film products is provided. In so doing so, the paper aims at contributing to the Eco-design of these innovative products.

1.1 The case study of mulch films

Plastic mulch films represent an important agronomical technique well established for the production of many crops thanks to numerous agronomical advantages such as: increased yield and higher quality of productions (Steinmetz et al., 2016) ; weed control and reduced use of pesticides; early crop production and reduced soil moisture loss (Briassoulis and Giannoulis, 2018). As a consequence, the plastic films consumption has increased year-by-year, reaching a current global market estimated at 1.4 millions of tonnes , mainly in Asia (Briassoulis and Giannoulis, 2018; Mormile et al., 2017) , and covering 80,000 km^2 of agricultural surface (0.6% of the global arable land). The mulch film market in Europe is estimated by Agriculture Plastic & Environment and by the European Bioplastic Associations at 76-80 kt. The most used raw material is Poly-Ethylene (PE) in its different forms, due to its processability, chemical resistance, high durability and flexibility (Kasirajan and Ngouajio, 2012; Plasticulture, 2016 and 2018; Shen, M. et al., 2019; Wen, X. et al., 2018).

Despite these benefits, manifold environmental and agronomic problems have been pointed out. After its useful life – which in general does not exceed 1 to 3 months – the mulch film has to be removed and properly disposed of, a time-consuming (about 16 hours per hectare) and costly procedure (Scaringelli, M., 2016; Briassoulis, D., 2013). The recovered film is usually heavily contaminated with soil and organic residues, making mechanical recycling technically difficult and not a cost-efficient solution (Briassoulis et al., 2018; Figuier, 2016; De Lèpinau, Arbenz, 2016). The most common end of life of collected films in Europe is still landfilling (about 50%), followed by energy recovering

and finally mechanical recycling (Le Moine, 2014). Recent Chinese prohibition (January 2018) to import different types of wastes is heavily impacting the European agricultural plastic waste management, highlighting the difficulty in properly recycling this type of plastics (Tamma, 2018). Plastic films may not be properly collected and recycled but disposed of by burning in the field or by uncontrolled landfilling or left directly in the (agricultural) soils, causing serious environmental concerns. An example is the “White pollution” phenomena described in the Xinjiang Autonomous Region (China), in which the residual plastic film can reach 200 kg/ha in the top soil with detrimental effects on soils’ quality, health and fertility (Liu, He, & Yan, 2014; Gao *et al.*, 2019; Steinmetz *et al.*, 2016).

As a reaction, there has been significant research into novel materials especially related to biodegradable and bio-based (BB) mulch films, which enable an effective biodegradation in soil and provide comparable agronomical performances (Touchaleaume et al., 2016). The term “bio-mulch film” brings together several types of both bio-based and fossil oil-based biodegradable polymers and blends of them, such as polylactic acid (PLA), polybutylene adipate co-terephthalate (PBAT), starch-based polymer blends or copolymers. They biodegrade when exposed to bioactive environments such as soil and compost (Kasirajan et al., 2012) which means that they can be left *in situ* to be fully biodegraded after being used. Clearly the biodegradation rate of biodegradable bioplastics is influenced by the environmental conditions such as the types of available bacteria, fungi thus specific enzymes namely native microflora (Pico, Y. et al., 2019). However their intrinsic biodegradability allow the complete biodegradation with times similar to natural polymers such as cellulose used as reference by the relevant standards and certification schemes.

The EN 17033:2018 is a new European Norm (standard) concerning “Plastics - Biodegradable mulch films for use in agriculture and horticulture - Requirements and test methods”, which sets the necessary tests and limits to define biodegradability, performances and environmental impacts of BB much films. The material is considered completely biodegradable if it achieves a complete biodegradation (absolute or relative to the reference material) in a test period no longer than 24 months (mineralization into CO_2). Additionally, a control of constituents (such as metals) and eco-toxicity testing (acute and chronic toxicity tests on plant growth, earthworm; nitrification inhibition test with soil microorganisms) were required. A certified mulch film guarantees that the product will completely biodegrade in the soil without adversely impacting on the environment.

1.2 Goal of the paper

The goal of the paper is to provide a general and common metric to measure the circularity of a bio-based and biodegradable (BB) product and to apply the methodology at product level to a category of products, namely bio-based and biodegradable mulch films.

2 Materials and Methods

2.1 MCI accounting according to the EMF methodology

The Material Circularity Indicator (MCI), according to the Ellen MacArthur Foundation (EMF) methodology (Ellen MacArthur Foundation & Granta Design, 2015), is a number that can range from 0 (pure linearity) to 1 (pure circularity). A purely linear production provides for the exclusive use of virgin raw materials that turn into waste at the end of the use phase of the product. Vice-versa, pure circularity includes the use of recycled materials and does not produce wastes (regenerative streams). Circularity can be achieved in different ways: as for the purpose of this paper, only recycling will be considered since

reuse is not an option for thin biodegradable mulch films. Since the method considers only mass flows, the recycling corresponds to the recovery of materials for the original purpose or for other purposes and excludes energy recovery, considered as a loss of materials equal to landfill disposal. The materials recovered feed back into the process as recycled feedstock.

The MCI methodology differentiates 'technical cycles' from 'biological cycles', modelling only the former. The first contains products and materials re-entering into the system (market) with the highest possible qualities and for as long as possible (thanks to reuse, repair, refurbishment and recycling) and the latter includes biological materials used in cascade until their restoration into the biosphere and the re-constitution of natural resources.

The material flows associated to the production of a generic technical cycle from non-renewable sources are summarized in Figure 1~~Figure 1~~. The dashed lines indicate that recycled feedstock does not have to be sourced from the same product but can be acquired on the market. With reference to Figure 1~~Figure 1~~, the list of the parameters used in the EMF methodology is reported in Table 1~~Table 1~~, while the equations relevant for the analysis carried out in this paper are described in the following sections (Table 2~~Table 2~~, Chapter 2.2).

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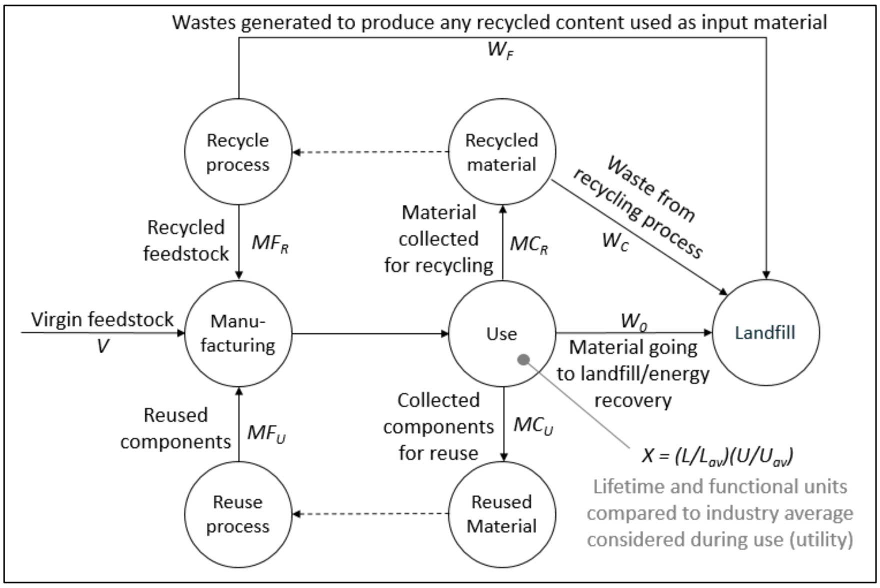

Figure 1: *Diagram of material flows and associated variables of a generic product (modified from Ellen MacArthur Foundation & Granta Design, 2015).*

Table 1: *Parameters and relative definitions used in the EMF methodology.*

Parameter	Definition
M	Total mass of the product
F_R	Fraction of mass of a product's feedstock from recycled sources
F_U	Fraction of mass of a product's feedstock from reused sources
V	Mass of virgin feedstock used in a product
C_R	Fraction of mass of a product being collected to go into a recycling process
C_U	Fraction of mass of a product going into component reuse
E_C	Efficiency of the recycling process used for the portion collected for recycling
E_F	Efficiency of the recycling process used to produce recycled feedstock for a product

W	Total mass of unrecoverable waste associated with a product
W_0	Mass of unrecoverable waste (landfill, waste to energy and any other type of process where the materials are no longer recoverable)
W_C	Mass of unrecoverable waste generated in the process of recycling parts of a product (after use)
W_F	Mass of unrecoverable waste generated when producing recycled feedstock for a product
X	Utility of a product, calculated as $X = (L/L_{av})(U/U_{av})$
L	Actual average lifetime of a product
L_{av}	Actual average lifetime of an industry-average product of the same type
U	Actual average number of functional units achieved during the use phase of a product
U_{av}	Actual average number of functional units achieved during the use phase of an industry-average product of the same type

The Material Circularity Indicator is determined as follows: , where *LFI* is the Linear Flow Index measuring the flows of virgin materials and unrecoverable wastes associated to the examined product.

A function of the utility, – , is used to correct the *LFI*. The function *F* is chosen in such a way that improvements of the utility of a product (e.g., by using it longer) have the same impact on its MCI as a reuse of components, leading to the same amount of reduction of virgin material use and unrecoverable waste. Setting $a = 0.9$, MCI takes, by convention, the value 0.1 for a fully linear product (*i.e.*, $LFI = 1$) whose utility equals the industry average (*i.e.*, $X = 1$). This leaves some margin to distinguish between processes with a high linearity but different utilities.

2.2 *MCI accounting for bio-based and biodegradable (BB) products*

To apply the EMF methodology to BB products, formulas and flows (Figure 1~~Figure 1~~ and Figure 2~~Figure 2~~) are adapted as it follows:

1. The fraction of the recycled feedstock, F_R, corresponds to the share of the bio-based feedstock content in the final BB product, $F_{R(i)}$. It is the ratio of the d.m. amount of bio-based feedstock per d.m. amount of the total mass of BB product (EN 16785-2:2016).
2. The fraction of restorative mass going into a recycling process, C_R, corresponds to the share of bio-based feedstock content in the BB product biologically recovered (*e.g.* through composting) or biodegraded in the natural environment, as it happens for specific applications (e.g. biodegradable mulch film, *etc.*). It is the ratio of the d.m. amount of bio-based feedstock per d.m. amount of the total mass of BB product that is biologically recycled.

The modified scheme is shown in Figure 2~~Figure 2~~. Table 2~~Table 2~~ lists the formulas as adapted to BB products.

Table 2: *List of formulas as developed by EMF methodology compared to the proposed adaptation to BB products.*

EMF methodology	**Adaptation to BB products**

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The mass of fossil-based feedstock which may be contained in BB products (V) is obtained as a difference of the total mass (M) minus the bio-based fraction; in this case the F_R in the EMF methodology corresponds to the sum of the fractions of all the bio-based feedstock/s used in manufacturing the BB product. Therefore, is the total bio-based feedstock mass in the product. In single-use products, such as mulch films, reuse is not considered for BB products, so that $F_U = C_U = 0$.

W_F is the total amount of unrecoverable waste associated to the production of bio-based feedstock used to produce BB products (*i.e.* the amount of uncoverable waste per unit of BB product). Bio-based feedstocks such as starch, PLA, PHB etc. generate non-restorative flows which can be quantified. Such unrecoverable waste correspond to $R_{(i)}$, the specific amount of waste generated within cradle-to-gate boundaries per unit of bio-based feedstock going into manufacturing, and it is estimated through LCA studies. Thus all inputs from growth and harvesting phases and the related wastes generated by fertilisers and pesticides are here accounted. $R_{(i)}$ can be easily found in specific literature or life cycle inventories (LCI) present in LCA databases. In the calculation of W_F, also the efficiency of manufacturing process of BB products E_P is considered, as the ratio of the

overall bio-based feedstock content in the final BB product to the bio-based feedstock in input to the manufacturing process.

The material flows associated to the production of a generic BB product are summarized in Figure 2~~Figure 2~~.

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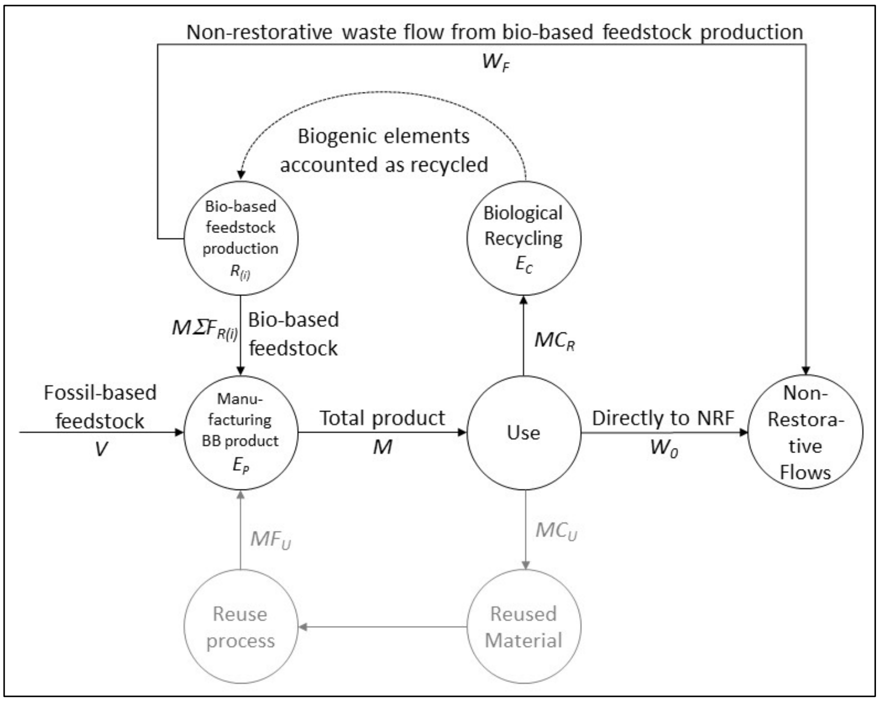

Figure 2: *Description of material flows adaptation to BB products; in this paper, the reuse flow is out of scope ($C_U = F_U = 0$).*

The biodegradation of bio-based feedstock does not imply the generation of waste W_C as it occurs in a standard mechanical recycling process. This implies that C_R and E_C (*i.e.* the efficiency of the biodegradation process) equal to 1. Indeed, a BB raw material, sent to biological treatment (composting) or biodegraded in a natural environment, is fully transformed in its chemical elements (C, H and O mainly) derived from the decomposition of complex molecules (polymers) without the release of waste (Witt et al., 2001; Marten et

al., 2003; Eubeler et al., 2010; BASF, 2018; Institute of Bioplastics and Biocomposites, 2018; OWS, 2018; Zumstein et al., 2018). These natural elements return into the environment and are then available in the respective biogeochemical cycles. The (biodegradable) fossil portion behaves as well; consequently, $W_C = 0$.

Nevertheless, the fossil-based feedstock cannot be considered as a regenerative circular feedstock, since it derives from carbon stored for millions of years and extracted by man, not being part of the active and fast biogeochemical carbon cycle. This is accounted in the quantification of W_0, the mass of unrecoverable waste from use (*i.e.* the linear stream going to landfill or incineration, the Non-Restorative Flows, NRF), as , the total amount of fossil-based feedstock.

Since W_F and W_C are associated to complete different processes and W_C is always equal zero, the double counting issue does not occur and the quantification of W and *LFI* is modified as reported in Table 2~~Table 2~~.

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2.3 MCI calculation for mulch films: scope, inventory and assumptions

The new formulas reported in Table 2~~Table 2~~ were applied to a single use product namely a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1.

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In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the BB film has been arbitrarily assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e. 23% of starch, $F_{(S)}$, and 7% of a bio-based additive, $F_{(BA)}$), while the rest was assumed to consist of fossil feedstock (

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Figure 3~~Figure 3~~). Since a generalized approach was used and no primary data were implemented, the information were extrapolated from literature (Institute of Bioplastics and Biocomposites, 2018); the main characteristics of the two examined products are presented in Table 3~~Table 3~~.

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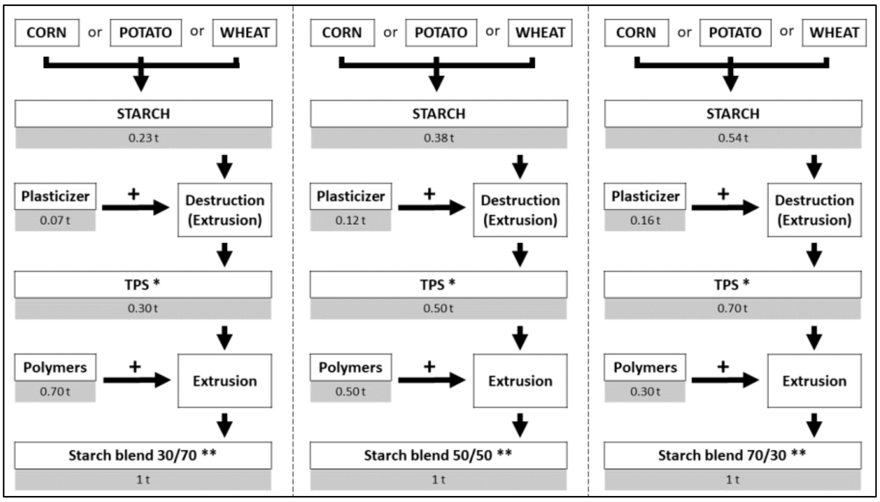

Figure 3: *Examples of hypothetical bio-based polymers; in this paper, the first option on the left (starch blend 30/70) has been chosen for carrying out the numerical MCI calculation (working hypothesis). The figure considers a 100%-efficiency in every phase of production, so that the residues are equal to zero; the same assumption is done in this paper. *TPS (Thermoplastic starch), starch content 75%; **Ratio TPS/Polymer; modified from Institute of Bioplastics and Biocomposites, 2018.*

Table 3: *Key features representative of the BB mulch films.*

	BB mulch film
Material	30% bio-based feedstock (23% starch + 7% bio-based additive) + 70% fossil feedstock
Thickness (µm)	12
Density (g/cm^3)	1.25
Weight (g/m^2)	15.2
Functional unit (the covering of the agricultural land)	6000 m^2/ha (the actual mulched soil in a hectare is generally equal to the 60% of the total area; Malinconico, 2017)

In the calculation of MCI for the BB mulch film, the adapted formulas were used together with assumptions. As stated before, BB mulch films are blends of bio-based and fossil based feedstocks (in the specific case, 30% and 70% respectively). Unlike the LDPE mulch film that has to be removed and disposed of, the BB mulch film is left in soil where it undergoes an ultimate biodegradation (so that $C_R = 1$) with no waste (so that $E_C = 1$), in respect of the specific standard EN 17033:2018. As a result of polymers' decomposition, the derived (biogenic) C, H and O finally return into biosphere (atmosphere, microorganism biomass, organic material pool) (OWS, 2018), and back into biogeochemical cycles in a relatively short time ("Biogenic elements accounted as recycled" in Figure 2~~Figure 2~~), with the exception of humified compounds. Actually, also C, H and O deriving from fossil-based sources undergo biodegradation (Zumstein, M.T., 2018) but they are not considered as a regenerative flow ("Waste from non-restorative flow" in *Figure 2*~~*Figure 2*~~) and their "wastes" are indeed calculated in W_0.

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Applying a conservative approach, W_F, the waste generated by the production of each bio-based feedstock, is quantified considering a "cradle to gate" LCA study. The estimated solid wastes $R_{(i)}$ for the presented case study are related to the production of starch ($F_{(S)}$),

with an amount $R_{(S)}$ of 0.014 kg of waste per kg of renewable feedstock (source: personal communication A. Novelli), and to the production of the bio-based additive ($F_{(BA)}$), with $R_{(BA)}$ equals to 0.025 kg waste/kg renewable feedstock (US-LCI database). As assumed in Figure 3Figure 3, the production efficiency of BB product E_P (how much bio-based feedstock is needed for every unit of BB product) is estimated equal to 1 and no unrecoverable wastes are generated by the process.

In addition, an explorative sensitivity analysis has been performed regarding exclusively the amount of bio-based feedstock content of the BB mulch film, *i.e.* (*i.e.*, $F_{(S)}$ + $F_{(BA)}$), as shown in Figure 4Figure 4 (Chapter 3). Considering the characteristics of the films (weight, g/m^2, or thickness, µm, and density, g/cm^3) and the relative functional unit (6000 m^2/ha, Table 3Table 3), it is possible to calculate a mass, *M*, that is 90 kg/ha for the BB one. Once calculated the masses, the formulas reported in Table 2Table 2 (Chapter 2.2) are applied. Results are shown in Table 4Table 4.

2.4 Sensitivity analysis

A sensitivity analysis was conducted for BB mulch film to examine the effects of changing the main variables. Given a non-linear dependence of results on parameter values, a Monte Carlo approach (see, *e.g.*, Lloyd and Ries, 2008) has been adopted. The model has been implemented using specifically written routines in the C++ programming language. The model was run with 100,000 events for BB mulch film, where the value of each parameter has been randomly chosen following a Gaussian distribution with a standard deviation within a range of possible and realistic values (Table 5Table 5 and **Error! Reference source not found.**Table 6; Figure 5Figure 5 and Figure 6Figure 6).

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3 Results

Figure 4~~Figure 4~~ shows how the value of the MCI varies according to the percentage variation of the bio-based feedstock in the total mass of the product.

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Table 4: *Resulting parameters in the calculation of MCI for BB mulch film.*

Parameter	BB mulch film

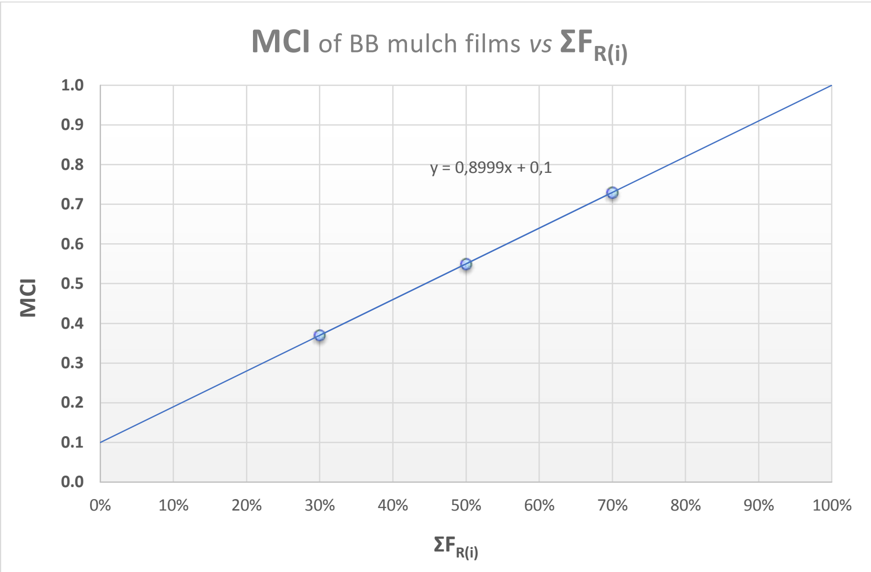

Figure 4: *MCI as a function of the amount of bio-based feedstock/s in the BB mulch film $\Sigma F_{R(i)}$, expressed as the percentage of all the bio-based feedstock/s of the mulch film on dry mass basis (X-axis). The dots correspond to the three different hypothetical bioplastic compositions of Figure 3.*

3.1 Sensitivity analysis

The results of the sensitivity analysis are presented in the followings Table 5~~Table 5~~ and Figure 5~~Figure 5~~ and Figure 6~~Figure 6~~. The accuracy band is a fraction of the average and corresponds to a probability of 95%. It has been chosen in order to be representative of the variability of the product category, the BB mulch films. The simulation can thus be regarded as a system composed by a high number of companies, each producing films with different characteristics, that are accounted for in the accuracy band.

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Table 5: *Parameters used for the sensitivity analysis of the BB mulch film. (**) The Accuracy Band is defined as twice the standard deviation of the distribution.*

Variable name	Average	Accuracy Band (**)	Unit
M	1000.00	0%	kg
$\mathbf{F_{(S)}/F_{(BA)}}$	3.29	10%	fraction
$\mathbf{F_{(S)} + F_{(BA)}}$	0.30	30%	fraction
$\mathbf{F_U}$	0.00	0%	fraction
$\mathbf{C_U}$	0.00	0%	fraction
$\mathbf{R_{(S)}}$	0.014	100%	fraction
$\mathbf{R_{(BA)}}$	0.025	100%	fraction
$\mathbf{E_C}$	1	0%	fraction
$\mathbf{E_P}$	0.95	10%	fraction
$\mathbf{C_R}$	1.00	0%	fraction

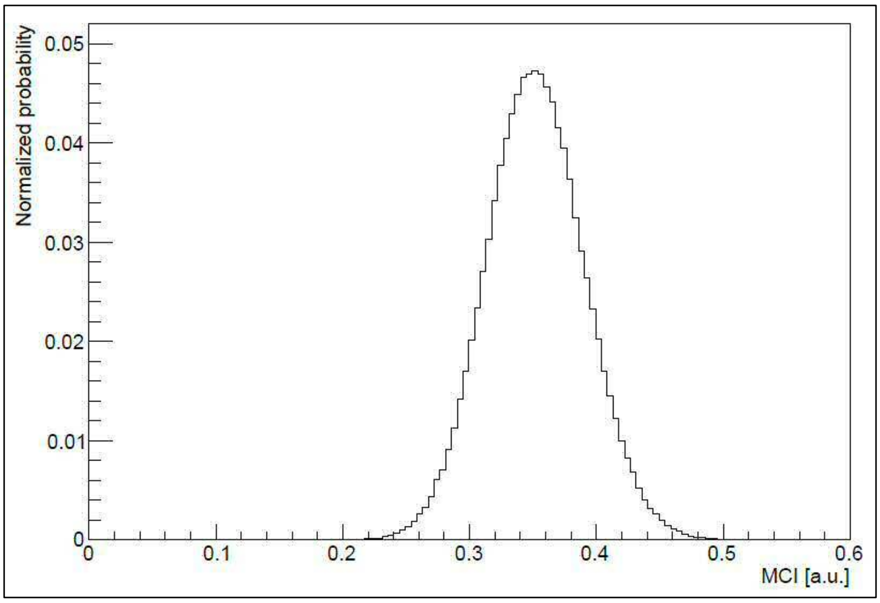

Figure 5: *Resulting distribution of MCI values for BB mulch film.*

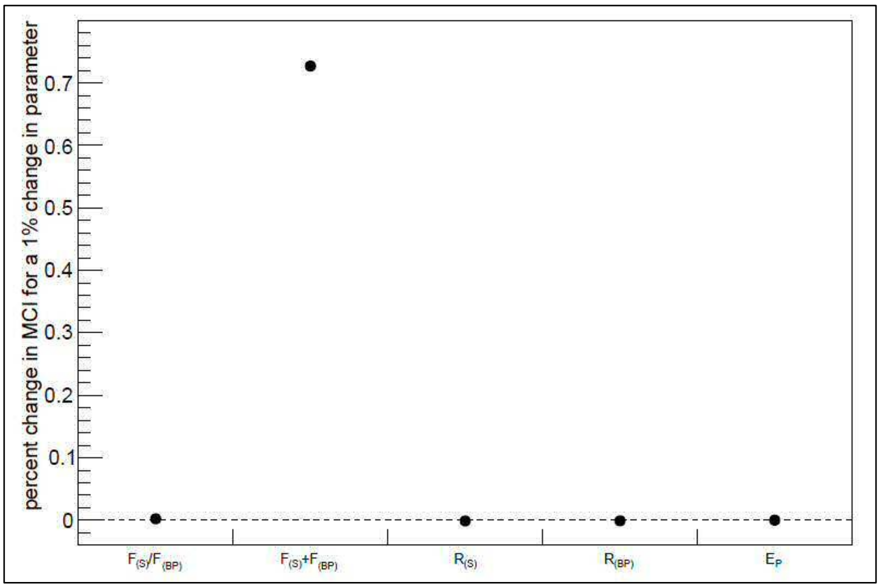

Figure 6: *The most sensitive and relevant parameters in the calculation of the MCI of the BB mulch films.*

4 Discussion

This work applies the principles of the EMF methodology into BB products so as to define common metrics for calculating their circularity. By doing so it proposes some substantial changes to the EMF methodology but still coherent with the overall methodological framework. Such changes should be seen as a generalisation of the methodology provided the following rules are applied:

(1) fossil-based feedstocks or component materials embodied in the BB products whatever is the final disposal (even biological recycling) shall be considered as non-restorative;

(2) bio-based component materials embodied in the BB product that go to biological recycling like composting, or biodegrade in the environment (i.e. BB mulch film) shall be considered restorative as long as they flow through the biosphere safely, without any harm to the environment (e.g. no toxicity effects).

(3) bio-based component materials embodied in the BB product that go to incineration and landfill shall be considered as non-restorative;

The justification of these rules is described in the following.

Fossil-based component materials in the product derive from deposits where they remained stocked for a geological time scale. Once the product is mineralised, its fossil-based portion will be accounted as non-regenerative and therefore linear, due to its origin (Joos et al., 2013). This is true, even if fossil carbon, for example, will re-enter biological cycles, like CO_2 in the atmosphere and other streams, since both fossil-based and bio-based component materials will physically and chemically behave the same, once biodegraded. However, the source of the bio-based carbon was circular before its use (concept of “carbon neutrality”, equilibrium between the biogenic carbon released and the carbon absorbed by plants) and will maintain its circularity provided that the carbon is released into the atmosphere at the same rate. The reason has its origin in the EMF general

provisions stating that "biologically sourced materials can only be considered part of a Circular Economy if materials are not used faster than they can be restored naturally" (Ellen MacArthur Foundation & Granta Design, 2015). If BB products are incinerated, the bio-based components are still considered linear, maintaining consistency with EMF principles. Basically, a complete circularity for a BB product is satisfied when its renewable components are 100% bio-based and they go 100% to biological recycling or biodegraded in the environment (for specific application like mulch film).

As for provision (3), a material health rule has its origin in mani~~y~~fold normative definitions of the CE. In addition, the EMF definition of biological cycles is that of non-toxic materials which are restored into the biosphere and the CE is defined as such if it can "eliminate the use of toxic chemicals". The need of a safety clause has been reviewed under many aspects by Verberne (2016) and can be put as a postulate of the restoration principle: if a flow is toxic it cannot be defined restorative. This is also at the core of the REACH Regulation (EC 1907/2006). In the specific case, the material complies with the standard EN 17033-2018 certifying that no harm is caused to a) all relevant organism groups as plants, invertebrates (e.g. earthworm) and microorganisms, b) important ecological processes maintaining soil functions, c) all relevant exposure pathways as soil pore water, soil pore air and soil material.

A comprehensive approach for MCI calculation should also include non-restorative flows generated at upstream level like biomass growth, in the specific case corn, and biomass conversion processes like starch extraction and refining. Specifically these non-restorative flows correspond to the overall non-recyclable wastes associated to the bio-based feedstock supply thus non-recyclable waste from fertilizer and pesticide production, non-recyclable scraps from conversion processes, etc. In this study such flows of non-restorative waste coming from upstream manufacturing operations were included for the

bio-based feedstocks ($R_{(i)}$) used in manufacturing the BB mulch film applying "cradle to gate" LCA methodology. However, we observed that the inclusion of upstream unrecoverable waste does not significantly influence the MCI results in the chosen case study, since the respective amounts are small. The specific unrecoverable waste for starch and bio-based additive (*i.e.* kg of waste/kg of bio-based feedstock) were estimated at 0.014 and 0.025, respectively.

The resulting MCI for the 30/70 blend of the BB mulch film is equal to 0.37 in a 0-1 scale and its circularity is linearly linked to the amount of bio-based feedstock used according to the equation $y = 0.89x + 0.1$, where y is the MCI and x is the bio-based feedstock content, therefore the amount of recycled feedstock or (renewable) bio-based feedstock in input is decisive.

Apart from the specific application analysed in this paper, the proposed MCI method can be easily applied and calculated for any kind of BB product as long as the following information are available:

- The bio-based feedstock content, determined according to the standard EN 16785-2:2016, if the composition is known, or directly provided by the BB product manufacturer.
- The end of life scenario of the studied BB product (real or hypothetical).
- The amount of un-recoverable waste associated to the production of bio-based feedstock contained in the BB product. They can be derived from LCA databases or other specific sources.

5 Conclusions

Bioplastic market is steadily increasing. The value proposition of bio-based and biodegradable products is linked to:

1. the use of renewable feedstock (like starch and its derivates) instead of fossil oil or natural gas;
2. the waste recovery through biological recycling, thanks to their ability to biodegrade in composting facilities or in soil (*e.g.* biodegradable mulch film).

The Material Circularity Indicator (MCI), developed by the EMF, is a metric for quantifying “how much” a product is circular (MCI = 0, fully-linear product; MCI = 1, completely circular product) thus it represents a valuable tool for product eco-design purposes. However, it focuses solely on technical materials, mechanically recycled or reused, leaving out bio-based feedstocks and related biological treatments such as composting. Without common metrics it is not possible to pursue concrete actions, to achieve measurable results and to provide unequivocal references for all products. This research work aims at filling this gap through the development of a methodology coherent with EMF MCI methodology but able to catch the specificities of bio-based and biodegradable products and provide metrics for those innovative products. Direct uses are: (i) supporting the eco-design of innovative bio-based products and (ii) comparing the MCI of BB products with MCI of traditional products (e.g. fossil based).

The proposed method has been applied to a real case study (*i.e.* biodegradable mulch film) providing quantitative metrics about its circularity. Specifically considering a bio-based feedstock content of 30%, the correspondent MCI is 0.37 in a 0-1 scale and its circularity is heavily linked to the bio-based feedstock content according to this relation: $MCI_{(BB\ mulch\ film)} = 0.89*bio\text{-}based\ feedstock + 0.1$.

The MCI is a key performance indicator to develop more circular products, in line with the Circular Economy principles like the use of renewable materials and the reduction of the amount of not recoverable waste. MCI will support the development of innovative products just based on these two important characteristics specific for each BB

product/application and end of life scenario. Bioeconomy, thus also BB products, can provide valuable insights in transforming the current (linear) economy in a more circular one, however, the way the biomass is produced, processed and BB products are produced are fundamental aspects to be properly assessed and monitored. This can be done using specific methodologies like LCA. Within this context the proposed MCI has to be seen as a complementary (quantitative) tool for further qualifying the sustainability of BB products and not as a substitute tool. Furthermore the MCI here proposed is meaningful only if BB products meet health and safety material requirements according to the national and European laws and standards. This is a postulate of the proposed methodology especially for those BB products conceived to biodegrade in the environment like biodegradable mulch film.

Declaration of interest

The author declares that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Addendum

While this paper was undergoing peer review the authors became aware that the EMF published an update of the MCI methodology (Ellen MacArthur Foundation & Granta Design, 2019) including the extension of it to include the treatment of biological materials. This update introduces new definitions and formulas. The authors believe that most of the changes regarding accounting are in the direction here proposed and that this study can contribute as an illustration on how the material circularity of a biological based material can be addressed in a real case study. Furthermore the authors would like to highlight that the proposed methodology started long before the EMF changes: specifically the original idea dated back to 2017 and a beta version of it - not as it is now - was presented in the middle of 2018 at the Italian Circular Economy Stakeholder Platform (ICESP www.icesp.it).

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Metrics for quantifying the circularity of bioplastics: the case of bio-based and biodegradable mulch films

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Abstract

The concept of circularity and its quantification through the Material Circularity Indicator (MCI) is well established for traditional plastic products. In this paper a methodological approach for calculating the circularity of bio-based and biodegradable (BB) products is proposed and applied to BB mulch films. BB products are different from traditional products in as much as they are sourced and regenerated (recycled) not through technical cycles but the biological loop. The suggested method is an adaptation of the MCI where two major changes were made: (i) the mass of the bio-based component corresponds to the recycled material in input and (ii) the mass of the bio-based component leaving the system through composting or biodegradation in soil is accounted as recycled. The modified MCI supports the eco-design of innovative BB products and allows for the comparison of their circularity taking into account the biological source and the expected end of life process such as biodegradation. To demonstrate the adaptation, the method has been applied to BB mulch films. Results showed that the MCI of a biodegradable mulch film, characterized by an average bio-based feedstock content of 30% is 0.37 ± 0.04 in a 0-1 scale. For BB mulch film, the amount of bio-based feedstock is the most sensitive factor and controls linearly the value of the MCI.

Keywords: circularity indicators, circular economy, bioplastics, biodegradable mulch film, bio-based product, biodegradation

Abbreviations	
BB	Biodegradable and bio-based
CE	Circular Economy
d.m.	Dry matter
EMF	Ellen MacArthur Foundation
LCA	Life Cycle Assessment
LDPE	Low-Density Poly-Ethylene
MCI	Material Circularity Indicator
NRF	Non-Restorative Flows
PBAT	Polybutylene adipate terephthalate
PE	Poly-Ethylene
PLA	Polylactic acid
PHB	Poly hydroxy butyrate

1 Introduction

To overcome today's unsustainable model of 'take-make-dispose' and its related risks such as hikes in raw material prices, pressures on the environment, shortage of global resources and waste sinks, a circular approach needs to be applied. It is a new regenerative economic view, based on a balance between economy, environment and society, a total resource efficiency and a Zero Emission Strategy that aims to maximize products value with zero, or minimal, environmental impact (Ghisellini et al., 2016) . Together with structural changes in environmental legislation, new logistics, technologies and sharing schemes, the Circular Economy (CE) approach which is regenerative by design, aims at closing materials loops, *i.e.* at reducing virgin materials input and waste output.

In December 2015, the European Commission developed an Action Plan for Circular Economy (European Commission, 2015), where plastic was considered a priority to be tackled. In January 2018, an *EU Plastic Strategy* (European Commission, 2018) was adopted, in order to react to the increasing environmental problems concerning plastic production, consumption, use and disposal along the same lines of the CE approach. Two fundamental steps to increase the circularity of different plastic products are (i) the abandonment of fossil fuels, *i.e.* currently 90% of the plastic is produced by virgin petroleum-based feedstock (Ellen MacArthur Foundation, 2017), and (ii) the development of easily recyclable products which are recycled. Today, in EU the share of plastics collected for recycling is 30% while the use of recycled plastics is just 6% (European Commission, 2018).

Biodegradable and bio-based (BB) plastics are spreading across markets (Institute for Bioplastics and Biocomposites, 2018) as a valid contribution to meet CE aims and principles. This is true as long as the supply of renewable raw materials, generally from agriculture, is based on a sustainable approach and the conversion processes along the

supply chain are efficient and highly integrated in a Life Cycle Assessment (LCA) perspective (EPLCA – European Platform on LCA). While traditional plastics can be mechanically recycled or incinerated with energy recovery, BB plastic products offer new recycling routes in waste management, due to their biodegradability. Organic recycling (through composting or anaerobic digestion) or in the case of specific applications such as agricultural mulch films, biodegradation in the environment, offer additional recovery options resulting in less wastes and less contamination of soil by plastic residues (Razza et al., 2012; Lange, B., 2016). An extensive literature review about the potentialities and benefits of renewable and compostable bioplastics, encompassing market perspective, applications, economic effects etc. can be found here: (BBIA; European Bioplastics).

Nevertheless, the research and development of innovative products, such as the BB products, implies the development of methodologies and metrics capable of measuring their circularity. Without this it is not possible to achieve measurable results and improving actions, as well as provide unequivocal references for comparisons of products of the same type/category. In 2015 the Material Circularity Indicator (MCI) was developed (Ellen MacArthur Foundation & Granta Design, 2015) which aims to quantify the regeneration of a product's material flow and is considered one of the few, among sixteen CE indexes suiting a micro-scale assessment of circularity at product or company level (Lonca et al., 2018). However, it focuses solely on technical cycles and recycled materials. Furthermore, recovery and recycling through the biological cycle offered by industrial composting, anaerobic digestion or biodegradation in natural environments are not considered as end of life options. In order to apply the MCI system to BB plastic products, the development of an enhanced methodology is necessary.

The approach proposed by the authors allows to quantify the circularity of BB plastic products and to make comparisons with equivalent traditional plastic products. To

demonstrate the applicability of the proposed method a computational example for mulch film products is provided. In so doing so, the paper aims at contributing to the Eco-design of these innovative products.

1.1 The case study of mulch films

Plastic mulch films represent an important agronomical technique well established for the production of many crops thanks to numerous agronomical advantages such as: increased yield and higher quality of productions (Steinmetz et al., 2016) ; weed control and reduced use of pesticides; early crop production and reduced soil moisture loss (Briassoulis and Giannoulis, 2018). As a consequence, the plastic films consumption has increased year-by-year, reaching a current global market estimated at 1.4 millions of tonnes , mainly in Asia (Briassoulis and Giannoulis, 2018; Mormile et al., 2017) , and covering 80,000 km^2 of agricultural surface (0.6% of the global arable land). The mulch film market in Europe is estimated by Agriculture Plastic & Environment and by the European Bioplastic Associations at 76-80 kt. The most used raw material is Poly-Ethylene (PE) in its different forms, due to its processability, chemical resistance, high durability and flexibility (Kasirajan and Ngouajio, 2012; Plasticulture, 2016 and 2018; Shen, M. et al., 2019; Wen, X. et al., 2018).

Despite these benefits, manifold environmental and agronomic problems have been pointed out. After its useful life – which in general does not exceed 1 to 3 months – the mulch film has to be removed and properly disposed of, a time-consuming (about 16 hours per hectare) and costly procedure (Scaringelli, M., 2016; Briassoulis, D., 2013). The recovered film is usually heavily contaminated with soil and organic residues, making mechanical recycling technically difficult and not a cost-efficient solution (Briassoulis et al., 2018; Figuier, 2016; De Lèpinau, Arbenz, 2016). The most common end of life of collected films in Europe is still landfilling (about 50%), followed by energy recovering

and finally mechanical recycling (Le Moine, 2014). Recent Chinese prohibition (January 2018) to import different types of wastes is heavily impacting the European agricultural plastic waste management, highlighting the difficulty in properly recycling this type of plastics (Tamma, 2018). Plastic films may not be properly collected and recycled but disposed of by burning in the field or by uncontrolled landfilling or left directly in the (agricultural) soils, causing serious environmental concerns. An example is the "White pollution" phenomena described in the Xinjiang Autonomous Region (China), in which the residual plastic film can reach 200 kg/ha in the top soil with detrimental effects on soils' quality, health and fertility (Liu, He, & Yan, 2014; Gao *et al.*, 2019; Steinmetz *et al.*, 2016).

As a reaction, there has been significant research into novel materials especially related to biodegradable and bio-based (BB) mulch films, which enable an effective biodegradation in soil and provide comparable agronomical performances (Touchaleaume et al., 2016). The term "bio-mulch film" brings together several types of both bio-based and fossil oil-based biodegradable polymers and blends of them, such as polylactic acid (PLA), polybutylene adipate co-terephthalate (PBAT), starch-based polymer blends or copolymers. They biodegrade when exposed to bioactive environments such as soil and compost (Kasirajan et al., 2012) which means that they can be left *in situ* to be fully biodegraded after being used. Clearly the biodegradation rate of biodegradable bioplastics is influenced by the environmental conditions such as the types of available bacteria, fungi thus specific enzymes namely native microflora (Pico, Y. et al., 2019). However their intrinsic biodegradability allow the complete biodegradation with times similar to natural polymers such as cellulose used as reference by the relevant standards and certification schemes.

The EN 17033:2018 is a new European Norm (standard) concerning "Plastics - Biodegradable mulch films for use in agriculture and horticulture - Requirements and test methods", which sets the necessary tests and limits to define biodegradability, performances and environmental impacts of BB much films. The material is considered completely biodegradable if it achieves a complete biodegradation (absolute or relative to the reference material) in a test period no longer than 24 months (mineralization into CO_2). Additionally, a control of constituents (such as metals) and eco-toxicity testing (acute and chronic toxicity tests on plant growth, earthworm; nitrification inhibition test with soil microorganisms) were required. A certified mulch film guarantees that the product will completely biodegrade in the soil without adversely impacting on the environment.

1.2 Goal of the paper

The goal of the paper is to provide a general and common metric to measure the circularity of a bio-based and biodegradable (BB) product and to apply the methodology at product level to a category of products, namely bio-based and biodegradable mulch films.

2 Materials and Methods

2.1 MCI accounting according to the EMF methodology

The Material Circularity Indicator (MCI), according to the Ellen MacArthur Foundation (EMF) methodology (Ellen MacArthur Foundation & Granta Design, 2015), is a number that can range from 0 (pure linearity) to 1 (pure circularity). A purely linear production provides for the exclusive use of virgin raw materials that turn into waste at the end of the use phase of the product. Vice-versa, pure circularity includes the use of recycled materials and does not produce wastes (regenerative streams). Circularity can be achieved in different ways: as for the purpose of this paper, only recycling will be considered since

reuse is not an option for thin biodegradable mulch films. Since the method considers only mass flows, the recycling corresponds to the recovery of materials for the original purpose or for other purposes and excludes energy recovery, considered as a loss of materials equal to landfill disposal. The materials recovered feed back into the process as recycled feedstock.

The MCI methodology differentiates 'technical cycles' from 'biological cycles', modelling only the former. The first contains products and materials re-entering into the system (market) with the highest possible qualities and for as long as possible (thanks to reuse, repair, refurbishment and recycling) and the latter includes biological materials used in cascade until their restoration into the biosphere and the re-constitution of natural resources.

The material flows associated to the production of a generic technical cycle from non-renewable sources are summarized in Figure 1. The dashed lines indicate that recycled feedstock does not have to be sourced from the same product but can be acquired on the market. With reference to Figure 1, the list of the parameters used in the EMF methodology is reported in Table 1, while the equations relevant for the analysis carried out in this paper are described in the following sections (Table 2, Chapter 2.2).

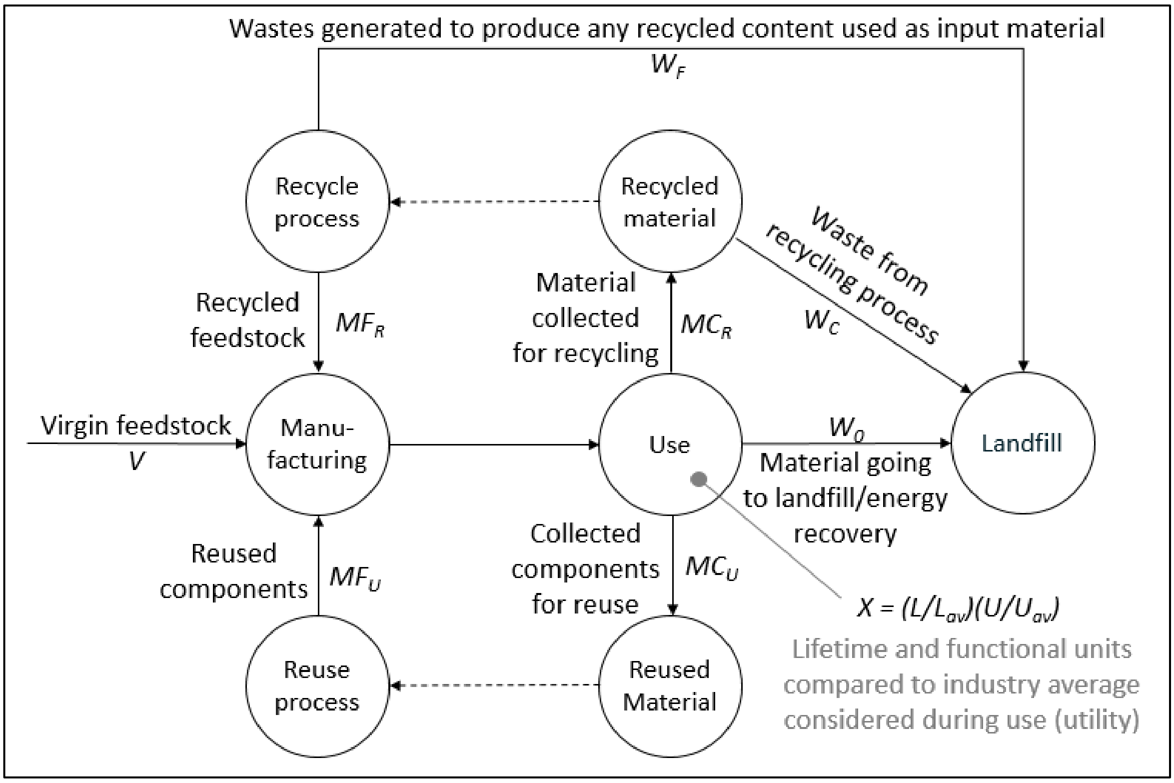

Figure 1: *Diagram of material flows and associated variables of a generic product (modified from Ellen MacArthur Foundation & Granta Design, 2015).*

Table 1: *Parameters and relative definitions used in the EMF methodology.*

Parameter	Definition
M	Total mass of the product
F_R	Fraction of mass of a product's feedstock from recycled sources
F_U	Fraction of mass of a product's feedstock from reused sources
V	Mass of virgin feedstock used in a product
C_R	Fraction of mass of a product being collected to go into a recycling process
C_U	Fraction of mass of a product going into component reuse
E_C	Efficiency of the recycling process used for the portion collected for recycling
E_F	Efficiency of the recycling process used to produce recycled feedstock for a product

W	Total mass of unrecoverable waste associated with a product
$\boldsymbol{W_0}$	Mass of unrecoverable waste (landfill, waste to energy and any other type of process where the materials are no longer recoverable)
$\boldsymbol{W_C}$	Mass of unrecoverable waste generated in the process of recycling parts of a product (after use)
$\boldsymbol{W_F}$	Mass of unrecoverable waste generated when producing recycled feedstock for a product
X	Utility of a product, calculated as $X = (L/L_{av})(U/U_{av})$
L	Actual average lifetime of a product
$\boldsymbol{L_{av}}$	Actual average lifetime of an industry-average product of the same type
U	Actual average number of functional units achieved during the use phase of a product
$\boldsymbol{U_{av}}$	Actual average number of functional units achieved during the use phase of an industry-average product of the same type

The Material Circularity Indicator is determined as follows: ,
where *LFI* is the Linear Flow Index measuring the flows of virgin materials and unrecoverable wastes associated to the examined product.

A function of the utility, − , is used to correct the *LFI*. The function *F* is chosen in such a way that improvements of the utility of a product (e.g., by using it longer) have the same impact on its MCI as a reuse of components, leading to the same amount of reduction of virgin material use and unrecoverable waste. Setting a = 0.9, MCI takes, by convention, the value 0.1 for a fully linear product (*i.e.*, $LFI = 1$) whose utility equals the industry average (*i.e.*, $X = 1$). This leaves some margin to distinguish between processes with a high linearity but different utilities.

2.2 MCI accounting for bio-based and biodegradable (BB) products

To apply the EMF methodology to BB products, formulas and flows (Figure 1 and Figure 2) are adapted as it follows:

1. The fraction of the recycled feedstock, F_R, corresponds to the share of the bio-based feedstock content in the final BB product, $F_{R(i)}$. It is the ratio of the d.m. amount of bio-based feedstock per d.m. amount of the total mass of BB product (EN 16785-2:2016).
2. The fraction of restorative mass going into a recycling process, C_R, corresponds to the share of bio-based feedstock content in the BB product biologically recovered (*e.g.* through composting) or biodegraded in the natural environment, as it happens for specific applications (e.g. biodegradable mulch film, *etc.*). It is the ratio of the d.m. amount of bio-based feedstock per d.m. amount of the total mass of BB product that is biologically recycled.

The modified scheme is shown in Figure 2. Table 2 lists the formulas as adapted to BB products.

Table 2: *List of formulas as developed by EMF methodology compared to the proposed adaptation to BB products.*

EMF methodology	Adaptation to BB products

The mass of fossil-based feedstock which may be contained in BB products (V) is obtained as a difference of the total mass (M) minus the bio-based fraction; in this case the F_R in the EMF methodology corresponds to the sum of the fractions of all the bio-based feedstock/s used in manufacturing the BB product. Therefore, is the total bio-based feedstock mass in the product. In single-use products, such as mulch films, reuse is not considered for BB products, so that $F_U = C_U = 0$.

W_F is the total amount of unrecoverable waste associated to the production of bio-based feedstock used to produce BB products (*i.e.* the amount of uncoverable waste per unit of BB product). Bio-based feedstocks such as starch, PLA, PHB etc. generate non-restorative flows which can be quantified. Such unrecoverable waste correspond to $R_{(i)}$, the specific amount of waste generated within cradle-to-gate boundaries per unit of bio-based feedstock going into manufacturing, and it is estimated through LCA studies. Thus all inputs from growth and harvesting phases and the related wastes generated by fertilisers and pesticides are here accounted. $R_{(i)}$ can be easily found in specific literature or life cycle inventories (LCI) present in LCA databases. In the calculation of W_F, also the efficiency of manufacturing process of BB products E_P is considered, as the ratio of the

overall bio-based feedstock content in the final BB product to the bio-based feedstock in input to the manufacturing process.

The material flows associated to the production of a generic BB product are summarized in Figure 2.

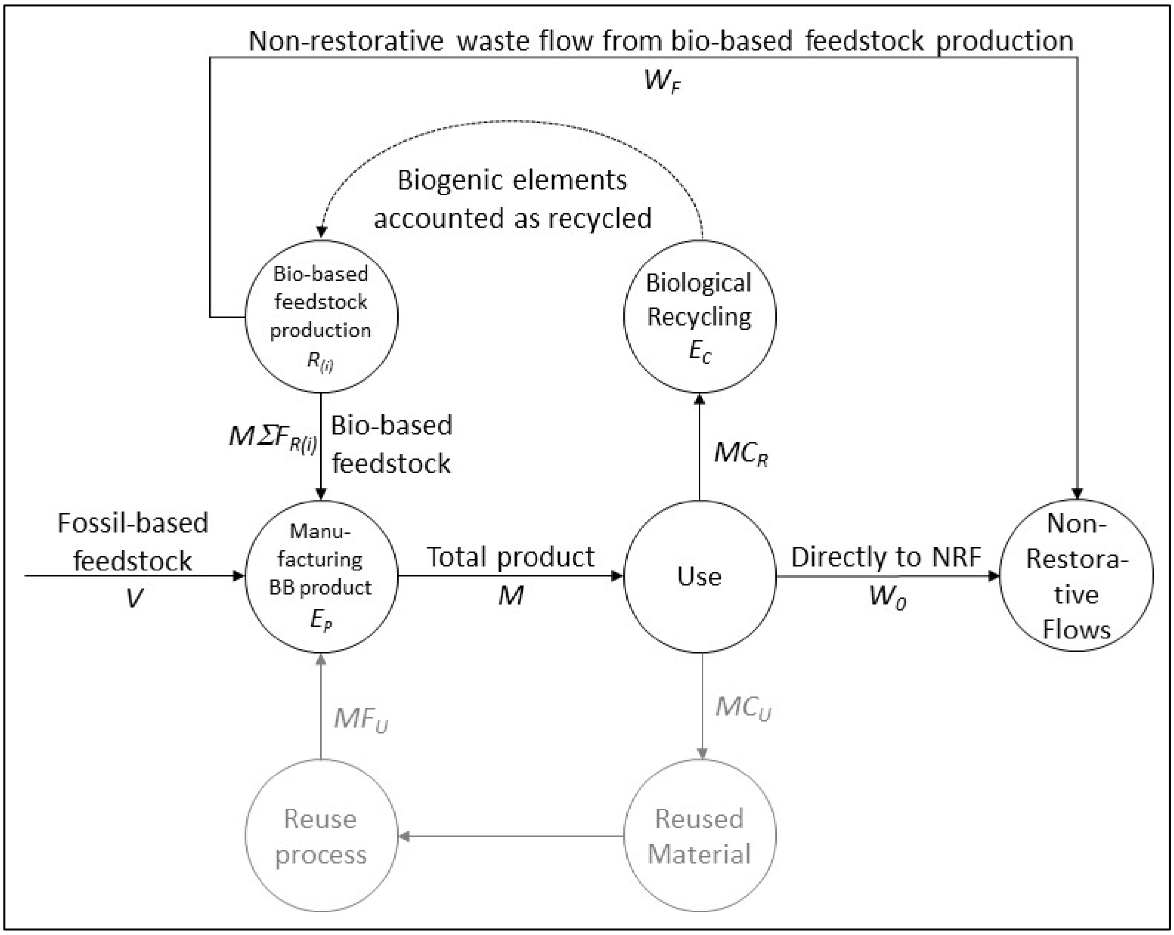

Figure 2: *Description of material flows adaptation to BB products; in this paper, the reuse flow is out of scope ($C_U = F_U = 0$).*

The biodegradation of bio-based feedstock does not imply the generation of waste W_C as it occurs in a standard mechanical recycling process. This implies that C_R and E_C (*i.e.* the efficiency of the biodegradation process) equal to 1. Indeed, a BB raw material, sent to biological treatment (composting) or biodegraded in a natural environment, is fully transformed in its chemical elements (C, H and O mainly) derived from the decomposition of complex molecules (polymers) without the release of waste (Witt et al., 2001; Marten et

al., 2003; Eubeler et al., 2010; BASF, 2018; Institute of Bioplastics and Biocomposites, 2018; OWS, 2018; Zumstein et al., 2018). These natural elements return into the environment and are then available in the respective biogeochemical cycles. The (biodegradable) fossil portion behaves as well; consequently, $W_C = 0$.

Nevertheless, the fossil-based feedstock cannot be considered as a regenerative circular feedstock, since it derives from carbon stored for millions of years and extracted by man, not being part of the active and fast biogeochemical carbon cycle. This is accounted in the quantification of W_0, the mass of unrecoverable waste from use (*i.e.* the linear stream going to landfill or incineration, the Non-Restorative Flows, NRF), as , the total amount of fossil-based feedstock.

Since W_F and W_C are associated to complete different processes and W_C is always equal zero, the double counting issue does not occur and the quantification of *W* and *LFI* is modified as reported in Table 2.

2.3 MCI calculation for mulch films: scope, inventory and assumptions

The new formulas reported in Table 2 were applied to a single use product namely a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1.

In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the BB film has been arbitrarily assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e. 23% of starch, $F_{(S)}$, and 7% of a bio-based additive, $F_{(BA)}$), while the rest was assumed to consist of fossil feedstock (

Figure 3). Since a generalized approach was used and no primary data were implemented, the information were extrapolated from literature (Institute of Bioplastics and Biocomposites, 2018); the main characteristics of the two examined products are presented in Table 3.

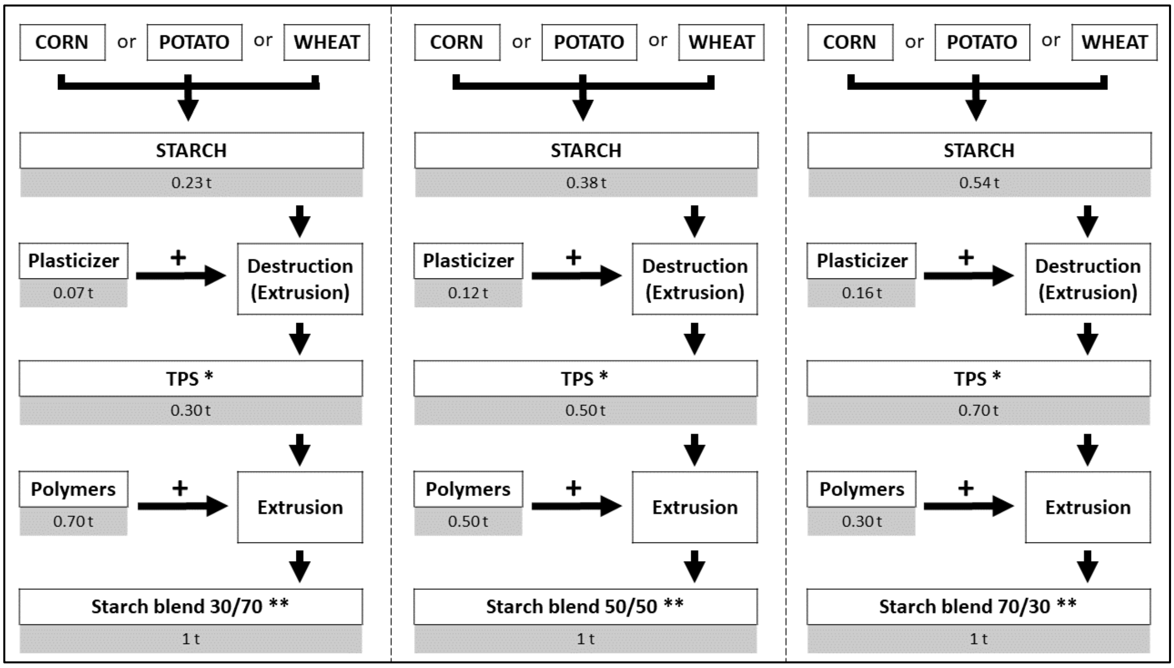

Figure 3: *Examples of hypothetical bio-based polymers; in this paper, the first option on the left (starch blend 30/70) has been chosen for carrying out the numerical MCI calculation (working hypothesis). The figure considers a 100%-efficiency in every phase of production, so that the residues are equal to zero; the same assumption is done in this paper. *TPS (Thermoplastic starch), starch content 75%; **Ratio TPS/Polymer; modified from Institute of Bioplastics and Biocomposites, 2018.*

***Table 3**: Key features representative of the BB mulch films.*

	BB mulch film
Material	30% bio-based feedstock (23% starch + 7% bio-based additive) + 70% fossil feedstock
Thickness (µm)	12
Density (g/cm^3)	1.25
Weight (g/m^2)	15.2
Functional unit (the covering of the agricultural land)	6000 m^2/ha (the actual mulched soil in a hectare is generally equal to the 60% of the total area; Malinconico, 2017)

In the calculation of MCI for the BB mulch film, the adapted formulas were used together with assumptions. As stated before, BB mulch films are blends of bio-based and fossil based feedstocks (in the specific case, 30% and 70% respectively). Unlike the LDPE mulch film that has to be removed and disposed of, the BB mulch film is left in soil where it undergoes an ultimate biodegradation (so that $C_R = 1$) with no waste (so that $E_C = 1$), in respect of the specific standard EN 17033:2018. As a result of polymers' decomposition, the derived (biogenic) C, H and O finally return into biosphere (atmosphere, microorganism biomass, organic material pool) (OWS, 2018), and back into biogeochemical cycles in a relatively short time ("Biogenic elements accounted as recycled" in Figure 2), with the exception of humified compounds. Actually, also C, H and O deriving from fossil-based sources undergo biodegradation (Zumstein, M.T., 2018) but they are not considered as a regenerative flow ("Waste from non-restorative flow" in *Figure 2*) and their "wastes" are indeed calculated in W_0.

Applying a conservative approach, W_F, the waste generated by the production of each bio-based feedstock, is quantified considering a "cradle to gate" LCA study. The estimated solid wastes $R_{(i)}$ for the presented case study are related to the production of starch ($F_{(S)}$),

with an amount $R_{(S)}$ of 0.014 kg of waste per kg of renewable feedstock (source: personal communication A. Novelli), and to the production of the bio-based additive ($F_{(BA)}$), with $R_{(BA)}$ equals to 0.025 kg waste/kg renewable feedstock (US-LCI database). As assumed in

Figure *3*, the production efficiency of BB product E_P (how much bio-based feedstock is needed for every unit of BB product) is estimated equal to 1 and no unrecoverable wastes are generated by the process.

In addition, an explorative sensitivity analysis has been performed regarding exclusively the amount of bio-based feedstock content of the BB mulch film, *i.e.* (*i.e.*, $F_{(S)}$ + $F_{(BA)}$), as shown in Figure 4 (Chapter 3). Considering the characteristics of the films (weight, g/m^2, or thickness, μm, and density, g/cm^3) and the relative functional unit (6000 m^2/ha, Table 3), it is possible to calculate a mass, *M*, that is 90 kg/ha for the BB one. Once calculated the masses, the formulas reported in Table 2 (Chapter 2.2) are applied. Results are shown in Table 4.

2.4 Sensitivity analysis

A sensitivity analysis was conducted for BB mulch film to examine the effects of changing the main variables. Given a non-linear dependence of results on parameter values, a Monte Carlo approach (see, *e.g.*, Lloyd and Ries, 2008) has been adopted. The model has been implemented using specifically written routines in the C++ programming language. The model was run with 100,000 events for BB mulch film, where the value of each parameter has been randomly chosen following a Gaussian distribution with a standard deviation within a range of possible and realistic values (Table 5 and **Error! Reference source not found.**; Figure 5 and Figure 6).

3 Results

Figure 4 shows how the value of the MCI varies according to the percentage variation of the bio-based feedstock in the total mass of the product.

Table 4: *Resulting parameters in the calculation of MCI for BB mulch film.*

Parameter	**BB mulch film**

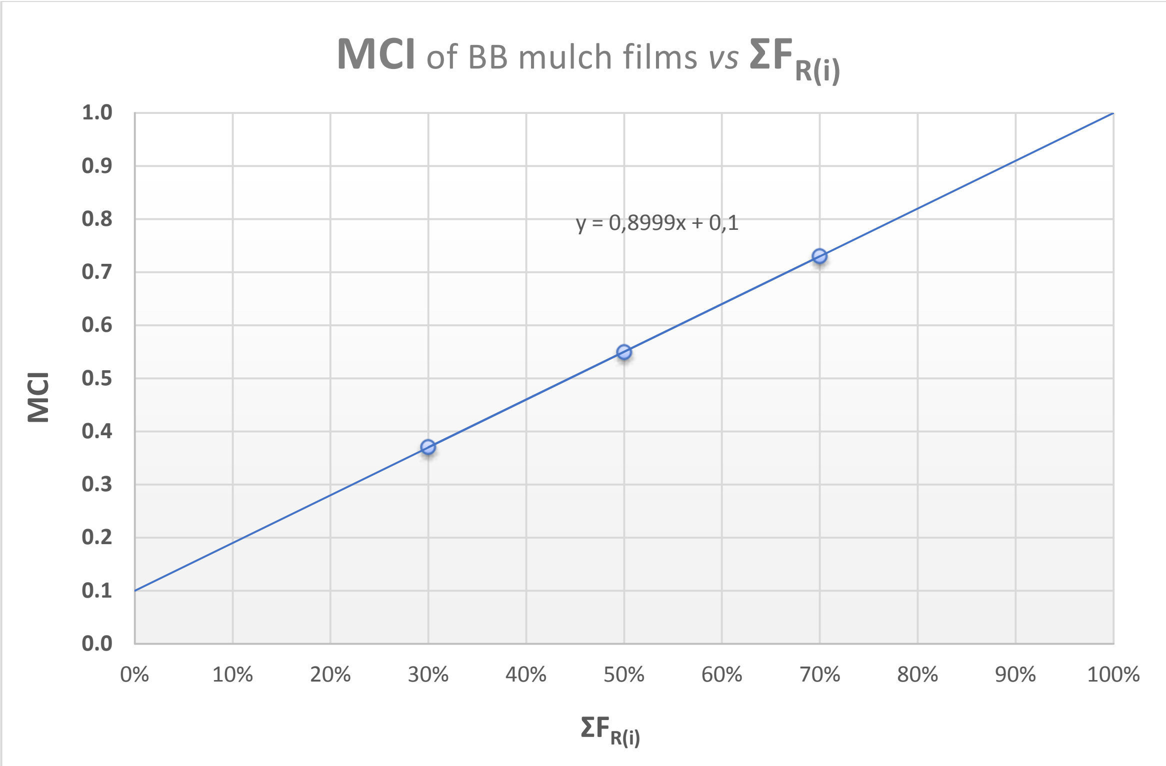

Figure 4: *MCI as a function of the amount of bio-based feedstock/s in the BB mulch film $\Sigma F_{R(i)}$, expressed as the percentage of all the bio-based feedstock/s of the mulch film on dry mass basis (X-axis). The dots correspond to the three different hypothetical bioplastic compositions of Figure 3.*

3.1 Sensitivity analysis

The results of the sensitivity analysis are presented in the followings Table 5 and Figure 5 and Figure 6. The accuracy band is a fraction of the average and corresponds to a probability of 95%. It has been chosen in order to be representative of the variability of the product category, the BB mulch films. The simulation can thus be regarded as a system composed by a high number of companies, each producing films with different characteristics, that are accounted for in the accuracy band.

Table 5: *Parameters used for the sensitivity analysis of the BB mulch film. (**) The Accuracy Band is defined as twice the standard deviation of the distribution.*

Variable name	Average	Accuracy Band (**)	Unit
$\mathbf{M}$	1000.00	0%	kg
$\mathbf{F_{(S)}/F_{(BA)}}$	3.29	10%	fraction
$\mathbf{F_{(S)} + F_{(BA)}}$	0.30	30%	fraction
$\mathbf{F_U}$	0.00	0%	fraction
$\mathbf{C_U}$	0.00	0%	fraction
$\mathbf{R_{(S)}}$	0.014	100%	fraction
$\mathbf{R_{(BA)}}$	0.025	100%	fraction
$\mathbf{E_C}$	1	0%	fraction
$\mathbf{E_P}$	0.95	10%	fraction
$\mathbf{C_R}$	1.00	0%	fraction

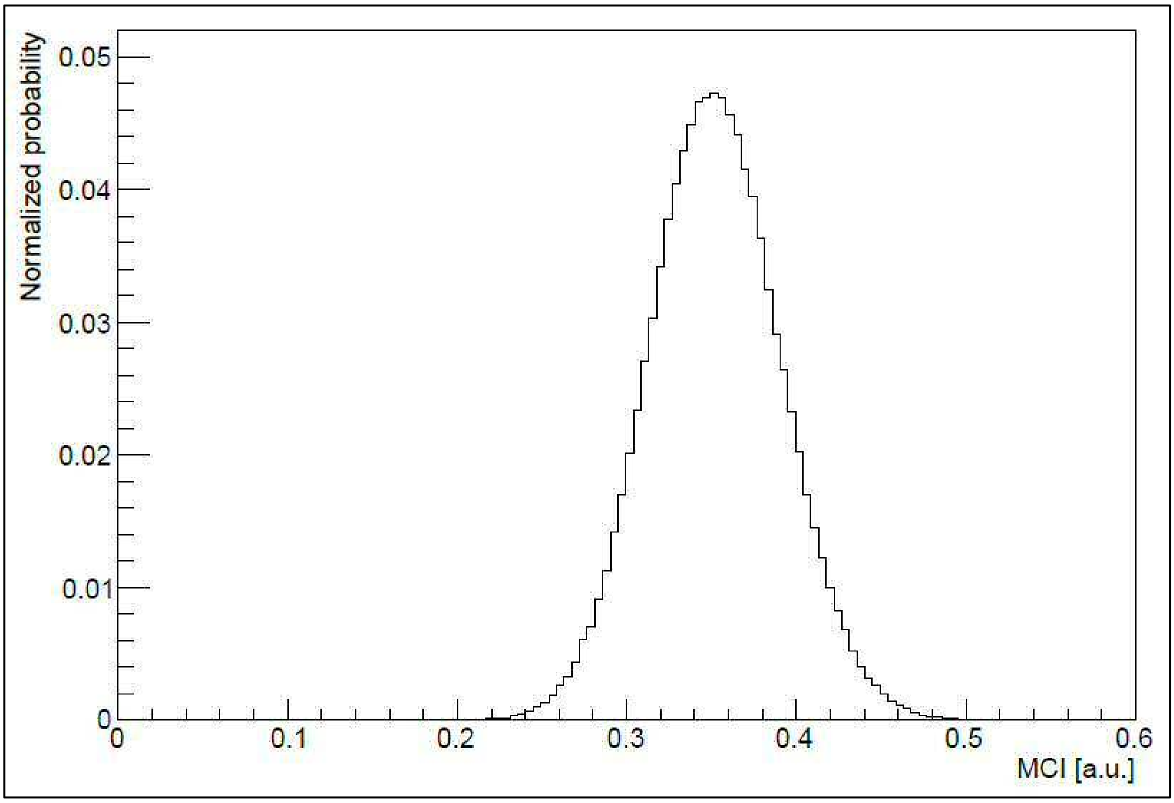

Figure 5: *Resulting distribution of MCI values for BB mulch film.*

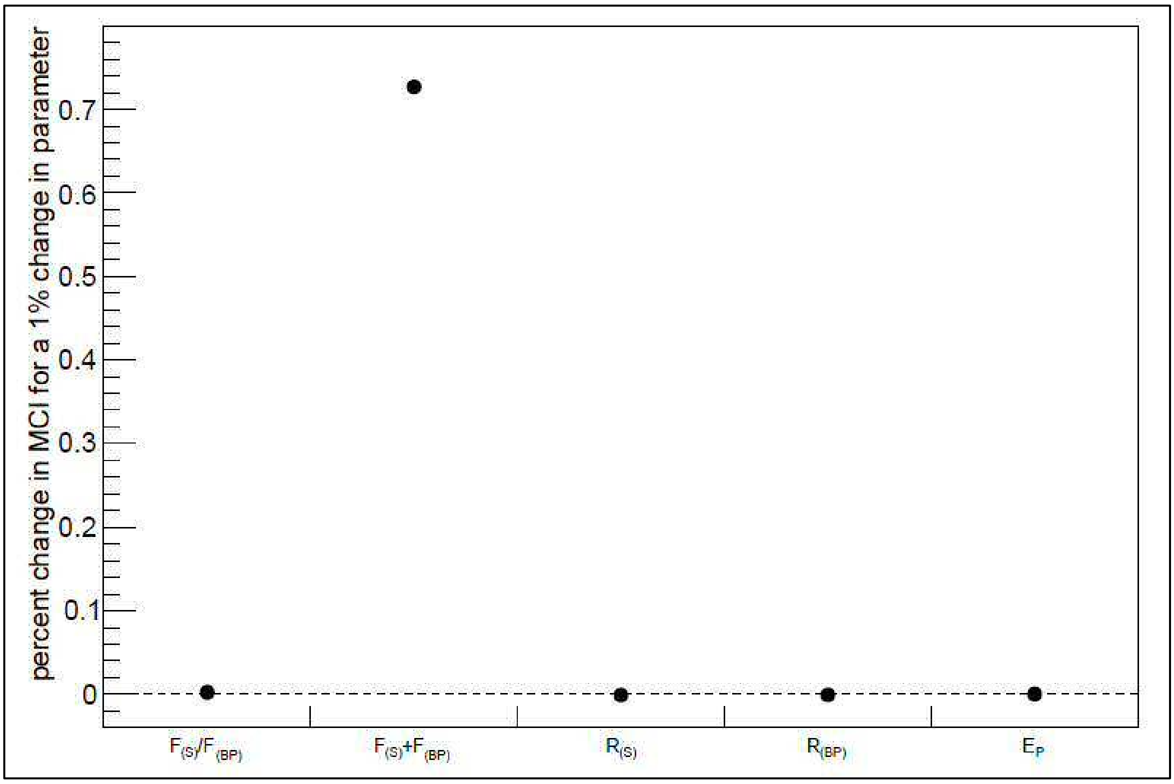

Figure 6: *The most sensitive and relevant parameters in the calculation of the MCI of the BB mulch films.*

4 Discussion

This work applies the principles of the EMF methodology into BB products so as to define common metrics for calculating their circularity. By doing so it proposes some substantial changes to the EMF methodology but still coherent with the overall methodological framework. Such changes should be seen as a generalisation of the methodology provided the following rules are applied:

(1) fossil-based feedstocks or component materials embodied in the BB products whatever is the final disposal (even biological recycling) shall be considered as non-restorative;

(2) bio-based component materials embodied in the BB product that go to biological recycling like composting, or biodegrade in the environment (i.e. BB mulch film) shall be considered restorative as long as they flow through the biosphere safely, without any harm to the environment (e.g. no toxicity effects).

(3) bio-based component materials embodied in the BB product that go to incineration and landfill shall be considered as non-restorative;

The justification of these rules is described in the following.

Fossil-based component materials in the product derive from deposits where they remained stocked for a geological time scale. Once the product is mineralised, its fossil-based portion will be accounted as non-regenerative and therefore linear, due to its origin (Joos et al., 2013). This is true, even if fossil carbon, for example, will re-enter biological cycles, like CO_2 in the atmosphere and other streams, since both fossil-based and bio-based component materials will physically and chemically behave the same, once biodegraded. However, the source of the bio-based carbon was circular before its use (concept of “carbon neutrality”, equilibrium between the biogenic carbon released and the carbon absorbed by plants) and will maintain its circularity provided that the carbon is released into the atmosphere at the same rate. The reason has its origin in the EMF general

provisions stating that "biologically sourced materials can only be considered part of a Circular Economy if materials are not used faster than they can be restored naturally" (Ellen MacArthur Foundation & Granta Design, 2015). If BB products are incinerated, the bio-based components are still considered linear, maintaining consistency with EMF principles. Basically, a complete circularity for a BB product is satisfied when its renewable components are 100% bio-based and they go 100% to biological recycling or biodegraded in the environment (for specific application like mulch film).

As for provision (3), a material health rule has its origin in manifold normative definitions of the CE. In addition, the EMF definition of biological cycles is that of non-toxic materials which are restored into the biosphere and the CE is defined as such if it can "eliminate the use of toxic chemicals". The need of a safety clause has been reviewed under many aspects by Verberne (2016) and can be put as a postulate of the restoration principle: if a flow is toxic it cannot be defined restorative. This is also at the core of the REACH Regulation (EC 1907/2006). In the specific case, the material complies with the standard EN 17033-2018 certifying that no harm is caused to a) all relevant organism groups as plants, invertebrates (e.g. earthworm) and microorganisms, b) important ecological processes maintaining soil functions, c) all relevant exposure pathways as soil pore water, soil pore air and soil material.

A comprehensive approach for MCI calculation should also include non-restorative flows generated at upstream level like biomass growth, in the specific case corn, and biomass conversion processes like starch extraction and refining. Specifically these non-restorative flows correspond to the overall non-recyclable wastes associated to the bio-based feedstock supply thus non-recyclable waste from fertilizer and pesticide production, non-recyclable scraps from conversion processes, etc. In this study such flows of non-restorative waste coming from upstream manufacturing operations were included for the

bio-based feedstocks ($R_{(i)}$) used in manufacturing the BB mulch film applying "cradle to gate" LCA methodology. However, we observed that the inclusion of upstream unrecoverable waste does not significantly influence the MCI results in the chosen case study, since the respective amounts are small. The specific unrecoverable waste for starch and bio-based additive (*i.e.* kg of waste/kg of bio-based feedstock) were estimated at 0.014 and 0.025, respectively.

The resulting MCI for the 30/70 blend of the BB mulch film is equal to 0.37 in a 0-1 scale and its circularity is linearly linked to the amount of bio-based feedstock used according to the equation $y = 0.89x + 0.1$, where y is the MCI and x is the bio-based feedstock content, therefore the amount of recycled feedstock or (renewable) bio-based feedstock in input is decisive.

Apart from the specific application analysed in this paper, the proposed MCI method can be easily applied and calculated for any kind of BB product as long as the following information are available:

- The bio-based feedstock content, determined according to the standard EN 16785-2:2016, if the composition is known, or directly provided by the BB product manufacturer.
- The end of life scenario of the studied BB product (real or hypothetical).
- The amount of un-recoverable waste associated to the production of bio-based feedstock contained in the BB product. They can be derived from LCA databases or other specific sources.

5 Conclusions

Bioplastic market is steadily increasing. The value proposition of bio-based and biodegradable products is linked to:

1. the use of renewable feedstock (like starch and its derivates) instead of fossil oil or natural gas;
2. the waste recovery through biological recycling, thanks to their ability to biodegrade in composting facilities or in soil (*e.g.* biodegradable mulch film).

The Material Circularity Indicator (MCI), developed by the EMF, is a metric for quantifying "how much" a product is circular (MCI = 0, fully-linear product; MCI = 1, completely circular product) thus it represents a valuable tool for product eco-design purposes. However, it focuses solely on technical materials, mechanically recycled or reused, leaving out bio-based feedstocks and related biological treatments such as composting. Without common metrics it is not possible to pursue concrete actions, to achieve measurable results and to provide unequivocal references for all products. This research work aims at filling this gap through the development of a methodology coherent with EMF MCI methodology but able to catch the specificities of bio-based and biodegradable products and provide metrics for those innovative products. Direct uses are: (i) supporting the eco-design of innovative bio-based products and (ii) comparing the MCI of BB products with MCI of traditional products (e.g. fossil based).

The proposed method has been applied to a real case study (*i.e.* biodegradable mulch film) providing quantitative metrics about its circularity. Specifically considering a bio-based feedstock content of 30%, the correspondent MCI is 0.37 in a 0-1 scale and its circularity is heavily linked to the bio-based feedstock content according to this relation: $MCI_{(BB\ mulch\ film)} = 0.89 * bio\text{-}based\ feedstock + 0.1$.

The MCI is a key performance indicator to develop more circular products, in line with the Circular Economy principles like the use of renewable materials and the reduction of the amount of not recoverable waste. MCI will support the development of innovative products just based on these two important characteristics specific for each BB

product/application and end of life scenario. Bioeconomy, thus also BB products, can provide valuable insights in transforming the current (linear) economy in a more circular one, however, the way the biomass is produced, processed and BB products are produced are fundamental aspects to be properly assessed and monitored. This can be done using specific methodologies like LCA. Within this context the proposed MCI has to be seen as a complementary (quantitative) tool for further qualifying the sustainability of BB products and not as a substitute tool. Furthermore the MCI here proposed is meaningful only if BB products meet health and safety material requirements according to the national and European laws and standards. This is a postulate of the proposed methodology especially for those BB products conceived to biodegrade in the environment like biodegradable mulch film.

Declaration of interest

The author declares that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Addendum

While this paper was undergoing peer review the authors became aware that the EMF published an update of the MCI methodology (Ellen MacArthur Foundation & Granta Design, 2019) including the extension of it to include the treatment of biological materials. This update introduces new definitions and formulas. The authors believe that most of the changes regarding accounting are in the direction here proposed and that this study can contribute as an illustration on how the material circularity of a biological based material can be addressed in a real case study. Furthermore the authors would like to highlight that the proposed methodology started long before the EMF changes: specifically the original idea dated back to 2017 and a beta version of it - not as it is now - was presented in the middle of 2018 at the Italian Circular Economy Stakeholder Platform (ICESP www.icesp.it).

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